Synchrotron Studies of X-ray Detectors

by

Anthony Peter Nichols

Thesis submitted to the University of Leicester for the degree of Doctor of Philosophy.

September 1996

X-Ray Astronomy Group
Department of Physics and Astronomy
University of Leicester
University Road
Leicester
U.K.
Abstract

This thesis is an account of research into various aspects of X-ray detection, with a common thread being the use of synchrotron radiation. An investigation into the so called "X-ray vectorial effect" is described. The aim being to design a detector able to determine the degree of polarisation in astronomical X-ray sources and the angle of the polarisation vector. Two experiments are reported, the first showing marked polarisation sensitivity, but not totally answering doubts. The second experiment demonstrated that the X-ray vectorial effect does not exist and with it the critical importance of accurate calibration of angles of incidence. Monte Carlo modelling of electron transport in photocathodes resulting from X-ray absorption is presented alongside compatible experimental results. An alternative polarimetry technique, also based on utilising the known polarisation sensitivity of photoelectron creation, is then derived and predicted to be competitive with the best current polarimeters and offering greater scope for improvement.

A novel efficiency calibration technique for bare Microchannel plates (MCPs) using the Daresbury synchrotron with very low beam current is examined allowing us to conduct photon counting measurements, while utilising the energy tunability of the synchrotron. In quantum efficiency measurements this technique brought out absorption edge fine structure and EXAFS from the constituents of MCP lead glass with the energy resolution required in calibration the MCP spectrometer readout on the AXAF satellite. Complimentary measurements of the quantum efficiency of alkali halide (CsI and KBr) photocathodes are presented over the continuous energy range between 2 and 8 keV and at a range of incidence angles. These alkali halides are generally coated onto the MCP channels to enhance the detector efficiency.

Synchrotron radiation was also utilised to examine the radiation damage characteristics of alkali halide photocathodes. Both CsI and KBr are shown to suffer significant degradation of photoelectric efficiency after minutes of irradiation with of order $10^{10}$ photons s$^{-1}$. The way X-ray flux, angle of incidence and energy affect the degree of degradation is investigated. A solid state model of lattice defect production, diffusion and trapping incorporating surface effects and photoelectron emission is described. This first attempt at modelling the physical basis of photoyield degradation is shown to accurately recreate much of the observed behaviour including the shapes of the signal decay curves and recovery out of the synchrotron beam yielding a clear insight into the degradation process and suggesting ways its effects can be reduced.
Declaration

I hereby declare that no part of this thesis has been previously submitted to this or any other University as part of the requirements for a higher degree. Work described here was conducted by the undersigned except for the contribution of colleagues indicated in the text.

[Signature]

Anthony P. Nichols
September 1996
Dedication

To my parents Keith and Susan Nichols,
To John Herod, Helen, Liz and Sid and
everyone else whose joie de vivre brightens all our lives.

Acknowledgements

I would especially like to thank my supervisor, Dr George Fraser for giving me the opportunity to spend such an enjoyable and fulfilling four years in the X-ray Astronomy group, for his enthusiasm, knowledgeable advice and for putting up with my sense of humour for so long. Special thanks also for the workers, Drs Jim Pearson, John Lees and Adam Brunton and those who got off with shorter sentences, Dr. Anton Tremsin, Sarah Pearce and Paul Houghton for many and varied stimulating discussions, occasionally related to the work presented in this thesis, and especially with others too numerous to mention, for their valuable contributions to the Daresbury experimental programmes. I would also like to thank all the Daresbury staff for helping us get the most out of our synchrotron time.

Dr Andrew Holland, Alex Short and George Fraser are fully deserved of my appreciation for their model of electron transport in silicon on which my multi-foil polarimeter design and model is based, as does Dr M. Marletta for his expert guidance on efficient computational methods for solving stiff systems of interdependent ordinary differential equations and for advising me that this was the type of problem I faced in modelling radiation damage in alkali halides.

I gratefully acknowledge the financial support of the Particle Physics and Astronomy Research Council (previously SERC) and Philips Photonics Ltd.
Publications Listing

Some of the experimental studies and theoretically analyses reported in this thesis have been incorporated in the following papers. The chapters to which these papers refer are given in brackets.

1. The X-ray Vectorial Effect revisited
   A. P. Nichols, J. F. Pearson, G. W. Fraser, J. E. Lees and M. Roper
   Nucl.Instr.Meth.B In preparation (Chapter 4)

2. The Primary electron vectorial effect and a promising new X-ray polarimeter
   A. P. Nichols and G. W. Fraser
   Nucl.Instr.Meth.B In preparation (Chapter 5)

3. Near Edge Structure in the Soft X-ray Quantum Efficiency of Microchannel Plate Detectors
   G. W. Fraser, J. E. Lees, J. F. Pearson, A. P. Nichols and P. Bailey

4. The Role of X-ray induced atomic defects in the degradation of photoelectric and scintillation yields from CaI, CaI(Tl) and KBr. A. P. Nichols and A. S. Tremsin
   Nucl.Instr.Meth.B In preparation (Chapters 8 and 9)
# Contents

1  The Daresbury Synchrotron Radiation Source (SRS) .............................................. 6  
   1.1  Synchrotron Radiation ........................................................................................... 6  
   1.2  The Daresbury SRS ............................................................................................... 8  
   1.3  Beamlines ............................................................................................................. 10  
      1.3.1  Beamline 6.1: Grazing Incidence Monochromator ....................................... 10  
      1.3.2  Beamline 6.3 .................................................................................................. 14  
      1.3.3  Beamline 3.4 .................................................................................................. 15  
      1.3.4  The Undulator Beamline 5U.1 ...................................................................... 17  
2  Astronomical X-ray Polarimetry .............................................................................. 22  
   2.1  Introduction .......................................................................................................... 22  
   2.2  Astrophysical Context ........................................................................................... 23  
   2.3  X-ray Polarimeters : General Principles ............................................................... 26  
      2.3.1  Bragg Crystal Polarimeter ............................................................................ 30
2.3.2 Thomson Scattering Polarimeter .............................................................. 31

2.3.3 The Stellar X-ray Polarimeter (SXRP) Instrument on Spectrum X-Gamma 33

2.3.4 Comparison of X-ray polarimeter designs ..................................................... 35

2.4 The X-ray Vectorial Effect .............................................................................. 36

2.4.1 Theoretical Considerations .......................................................................... 37

2.4.2 Previous Observations ................................................................................. 38

3 Polarimetric Measurements in the Energy Range 2-6 keV 50

3.1 Experimental Details ..................................................................................... 50

3.2 Gold Photocathodes ......................................................................................... 53

3.3 Caesium Iodide Photocathodes ....................................................................... 61

3.4 Discussion ........................................................................................................ 64

4 Polarimetric Measurements in the Energy Range 50 to 1000 eV 66

4.1 Experimental Details ..................................................................................... 66

4.2 Measurement Technique .................................................................................. 69

4.3 Total Electron Yields ..................................................................................... 70

4.4 Electron Energy Resolution ............................................................................. 80

5 X-ray Vectorial Effect Modelling 81

5.1 Polarisation Sensitive Primary Photoemission Model ................................... 81
Chapter 1

The Daresbury Synchrotron Radiation Source (SRS)

1.1 Synchrotron Radiation

Synchrotron Radiation has been described in great detail elsewhere (Kunz 1979a; Winick & Doniach 1980; Eastman & Farges 1983; Koch, Sasaki & Winick 1987), so I shall limit this discussion to the basics and to the properties that have made synchrotron radiation useful for the study of X-ray detectors. When any charged particle travels in a circular orbit it is radially accelerated, resulting in the characteristic radiation patterns seen in figure 1.1. For a low energy charged particle, the radiation emission profile is toroidal (Fig 1.1a). If, however, the particles orbit with a relativistic energy, the radiation is emitted in a narrow cone tangential to the orbital path (Fig 1.1b) and is almost completely polarised in the orbital plane. In practice, synchrotron rings use only the lightest charged particles, electrons and positrons, which can be accelerated near enough to the speed of light, c, to give off radiation of maximum intensity.

In synchrotrons and storage rings, powerful dipole magnets force the electrons to travel in sharply curved trajectories, then they travel straight for a while before reaching another bending magnet. Thus, the electron path resembles more a polygon than a circle and the synchrotron radiation is concentrated at the polygon corners rather than being circumferentially continuous. The
Figure 1.1: Angular intensity distributions of slow (a) and relativistic (b) electrons in a circular orbit (Tomboulian & Hartman 1956). $\beta$ is the electron velocity divided by the speed of light.
charged particles emit a continuum of synchrotron radiation whose spectral range depends on the energy of the particles and the field strength of the bending magnets. Synchrotron radiation is highly collimated, linearly polarised and is generally over three orders of magnitude more brilliant (photons per second per unit area per unit solid angle) than conventional laboratory light sources, as shown in figure 1.2. A typical spectrum from a synchrotron bending magnet is that marked NSLS bend (from the National Synchrotron Light Source at Brookhaven). The various curves at higher spectral brightnesses are from undulators and wiggler devices inserted in straight sections of the electron path. A description of insertion devices is given in section 1.3.4.

All of the experimental work presented in this thesis was carried out at the Synchrotron Radiation Source (SRS) at the CLRC Daresbury Laboratory. Section 1.2 describes this source; while section 1.3 describes the various beamlines (6.1, 6.3, and 3.4) located off the various SRS bending magnets. Beamline 5U.1, described in section 1.3.4, uses another type of magnet, an undulator (Suller 1980) as a radiation source. An undulator is an array of magnets on a straight section of the electron path, which produces radiation with very special features including a highly energy dependent degree of linear polarisation.

1.2 The Daresbury SRS

The Synchrotron Radiation Source (SRS) at the Daresbury Laboratory was designed and built in the late 1970s to give UK research workers access to the unique properties of synchrotron radiation. The useful spectral range runs continuously from the infra-red to the X-ray (10⁻⁴ eV to 100 keV) and dedicated experimental stations are provided to deliver specific energy bands to a wide range of user experiments. Figure 1.3 shows the layout of the Daresbury Laboratory. Electrons are produced and initially accelerated in the Linear Accelerator (Linac at the top centre of the diagram) before storage, further acceleration and addition of other packets of electrons in the booster. Electrons then pass to the 30m diameter main storage ring where the 16 bending magnets are labelled. Their path is so well defined by the magnets that electron lifetimes regularly reach 40 hours. Beam parameters for normal multi-bunch operation are typically 250 mA circulating current, with a beam energy of 2.0 GeV. The radiation is pulsed.
Figure 1.2: Spectral brightness of several synchrotron sources and conventional X-ray sources (Vaughan 1986)
with a pulse width of approximately 180 ps at a repetition rate of 500 MHz. The radiation is emitted from a very small area (≈ 0.8 mm²) and highly collimated, with a vertical divergence of order 1 milliradian.

1.3 Beamlines

A number of beamlines covering the energy range 50 eV to 10 keV have been used for the polarimetric and quantum efficiency measurements presented in this thesis. Beamline 6.1 covers the lowest part of this energy range and was utilised for our first polarimetric measurements (Section 2.4.2) and the Microchannel Plate calibration work (Chapter 6). Beamline 3.4 covers the energy range from 800 to 3000 eV and has been used for photocathode quantum efficiency experiments (Chapter 7) and radiation damage measurements (Chapter 8). The highest energy station used is line 6.3, is capable of covering the range 60 eV to 11.1 keV, but has only been utilised between 2 to 6 keV for measurements of photocathode polarimetric response (Chapter 3), alkali halide quantum efficiency determinations (Chapter 7) and radiation damage effects (Chapter 8). The special polarisation characteristics of beamline 5U.1 have been utilised in our most accurate X-ray polarimetric measurements to date, described in chapter 4.

1.3.1 Beamline 6.1: Grazing Incidence Monochromator

Beamline 6.1 is one of three stations off bending magnet 6. A mirror box just off the storage ring splits the beam into 3 components, one for each experimental station. A system of fast action valves along the beamlines ensure the integrity of the storage ring vacuum. The experimental arrangement of 6.1 is shown schematically in Figure 1.4.

The grazing incidence monochromator has been described by Howells et al. (1978) and is based on the original geometry of Miyake, Kato & Yamashita (1969). A gold coated plane grating made from fused quartz and a platinum coated Silicon Carbide spherical mirror monochromatise and reject high spectral orders from the beam. By selection of appropriate pre-mirror four overlapping energy regions between 40 and 300 eV can be accessed. However the change-over is complicated and generally only one range is used for a series of measurements. Range 2 produces
Figure 1.3: Plan of the Daresbury SRS
Figure 1.4: The grazing incidence monochromator on Beamline 6.1 at the Daresbury SRS.
the highest flux and thus was used for the experiments described in this thesis. This produces usable photon flux in the energy range 30 - 220 eV, peaking at approximately 150 eV. After exiting the monochromator the beam is defined by post monochromator slits which determine the beamline resolution. The beam is then focused at the centre of the experimental chamber by a gold coated ellipsoidal mirror. This results in a typical beam intensity at the sample (in our cases a microchannel plate or deposition photocathode) of \(10^{11}\) photons s\(^{-1}\) with a 0.3% bandwidth and a spot size of 1mm vertically and 3mm horizontally as illustrated in the KBr coated microchannel plate image, obtained during 'low ring current' operation of the SRS in January 1995 and shown in Figure 1.5.

The experimental chamber is pumped by an ion pump, a turbomolecular pump and a liquid
1.3.2 Beamline 6.3

Station 6.3 is the third of the three stations comprising line 6, designed primarily for surface EXAFS (Extended X-ray Absorption Fine Structure) study. Monochromatic photons are available with energies from 60 to 11000 eV, such a large range being covered by a choice of three double crystal monochromators for energies above 1745 eV and a plane grating/mirror monochromator for energies below 2000 eV. A schematic of the line in double crystal mode, as used for the experiments described herein, is shown in figure 1.6. The beam first passes through one of the available filters to remove ultra violet light that could otherwise specularly reflect of the monochromator crystals and thus be transmitted by the monochromator. The beam size is then defined by an adjustable aperture before being focused by the toroidal mirror. The mirror is a fused quartz crystal, coated with gold and bent into the toroidal shape and provides the high energy cut off of approximately 10 keV. Monochromatisation is achieved by a double crystal monochromator, consisting of three pairs of crystals (Ge(111), Ge(220) and Si(111)) mounted on a linear drive. By use of the drive the required crystal pair can be brought into the beam, Ge(111) \(2d = 6.532\,\text{Å}\) being used for the energy range 2000 - 8430 eV, Si(111) \(2d = 6.271\,\text{Å}\) between 2100 and 6000 eV and Ge(220) \(2d = 4.00\,\text{Å}\) for between 3260 and 11100 eV.

Conversion to grating mode is achieved by releasing the bend in the toroidal mirror so a parallel beam reaches a plane grating stored on the same linear drive as the monochromator crystals. Radiation is then focused by a spherical mirror to the sample.

Figure 1.7 shows a schematic of the beam monitoring section after the monochromator. The monochromator exit slits are left wide open in double crystal mode (used only for grating monochromator). Beam monitoring is achieved by two photodiode detectors \(I_{0}'\) and \(I_0\) separated by a rack of beam defining apertures. The first photodiode \(I_0'\) is made of 86 \% transmittance mesh, the drain current signal being directly measured by a picoammeter whose output is connected to a voltage to frequency converter and then to a CAMAC scaler. Above 2000 eV the 0.75 \(\mu\text{m}\) aluminium foil is generally used as the \(I_0\) monitor. The foils and meshes can be rotated to allow cleaning by the ion gun. This arrangement produces a focused spot size at the
sample of 4mm wide by 1mm high from a source region 6mm by 1.2mm.

1.3.3 Beamline 3.4

The experimental configuration for a photo-yield measurement on beamline 3.4 is shown in figure 1.8. The beam from the synchrotron ring is extracted from bending magnet 3 and reflected off a chromium flat, providing the high energy cut off, onto a toroidal mirror. The mirror focuses the beam onto the double crystal monochromator providing a collimated monochromatic beam at the sample. Five different crystal pairs, Beryl (2d = 15.954), Quartz (2d = 8.512), InSb(111) (2d = 7.4806), Si(111) and Ge(111)) are used to cover the available energy range of 800eV to 3500eV. These are interchangeable, with only one pair being installed at any one time. The beamline station consists of two experimental chambers with the beam focus lying mid way between the two to give both chambers a reasonably focused beam. The front chamber was used for the work presented in Chapters 7 and 8. The rear chamber being designed for reflected extended X-ray absorption fine structure (reflexafs) measurements.
Calibration foils/filters N
Double crystal monochromator
86% Transmission copper mesh
I₀ monitor
Leak valve
Ion gun Argon or oxygen
To sample
2.5µ Beryllium foil
0.75µ Aluminium foil
86% Transmission copper mesh
92% Transmission tungsten mesh
Pico ammeter
Monoctiromator exit slits wide open
Pico ammeter
Beam defining apertures

Figure 1.7: Beam Defining and monitoring section of beamline 6.3 (MacDowell, Norman & West 1986)

Figure 1.8: Experimental Arrangement for a photocurrent measurement on Beamline 3.4 at the Daresbury SRS. S1 and S2 are beam defining slits (SRS handbook 1991)
1.3.4 The Undulator Beamline 5U.1

Undulators and wigglers are magnetic devices inserted into straight sections of the synchrotron "ring" to produce radiation with distinctive properties for the Synchrotron radiation users. An undulator is really a special type of wiggler; both are arrays of dipole magnets arranged (for an undulator, figure 1.9) in such a way that the magnetic field is in the vertical direction and varies sinusoidally. The electron trajectory is then also sinusoidal and lies in the horizontal plane. An important parameter characterising insertion devices is the deflection parameter $K$ given by,

$$K = 0.934\lambda_0\beta_0$$  \hspace{1cm} (1.1)

where $\lambda_0$ is the length of the magnetic period in cm and $\beta_0$ the peak magnetic field in Tesla.

For wigglers $K$ is large and radiation from different parts of the electron trajectory sum incoherently. Every point on the electron path emits a spectrum similar to that of the bending magnet with critical wavelength similarly given by,

$$\lambda_c = \frac{18.6}{E^2\beta}$$  \hspace{1cm} (1.2)

where $E$ is the electron beam energy in GeV. With the magnetic field strength, $\beta$, for superconducting wiggler magnets up to 5 times those of bending magnets, wigglers can thus be used to
obtain higher X-ray energies without resorting to the far more expensive solution of increasing $E$.

Undulators, on the other hand, operate with a low value of $K$. The radiation is best described by considering the relativistic oscillation of the electron transverse to the electron beam motion. In this situation the electrons radiate at the oscillation wavelength, which in the laboratory frame, as given by Koch, Sasaki & Winick (1987) is:

$$\lambda = \frac{\lambda_0}{2\gamma^2 \left(1 + \frac{1}{2} K^2 + \gamma^2 \theta^2\right)} \quad (1.3)$$

where $\theta$ is the angle between the axis and the observational direction and $\gamma$ the relativistic factor equal to $(1 - \frac{v^2}{c^2})^{-\frac{1}{2}}$. For higher magnetic fields, the transverse force on the electron is increased, and allowing the electron to possess a relativistic transverse motion superimposed on its relativistic forward motion. Under these conditions any increase in transverse momentum must be accompanied by a corresponding decrease in longitudinal momentum and vice versa. Thus, from a moving frame of reference of the average velocity along the longitudinal axis, the electron executes a figure of eight type motion instead of the simple harmonic oscillation previously assumed. The effect of this is to add harmonics of the oscillation frequency, such that transformed to the laboratory frame the wavelength spectrum becomes,

$$\lambda = \frac{\lambda_0}{2\gamma^2 \left(1 + \frac{1}{2} K^2 + \gamma^2 \theta^2\right)} \quad (i = 1, 2, 3\ldots) \quad (1.4)$$

with only the odd harmonics ($i=1,3,5\ldots$) producing radiation in the plane of the electron oscillation. For an on axis detector, in terms of photon ($\epsilon$) and electron ($E$) energy rather than wavelength and gamma factor, this becomes;

$$\epsilon_1 = 950 \frac{E^5}{(1 + \frac{1}{2} K^2)\lambda_0} \quad (1.5)$$

Further increases in magnetic field smooth out the harmonic structure to reproduce the continuous spectra of the wiggles.
The total power output from an undulator is moderate (~ 20 - 30 W) although at the fundamental energy, the brilliance (photons/mm²/mrad²/s/unit bandwidth) is 10⁴ times that from a bending magnet. It is for this reason undulators are generally commissioned and used.

The undulator source of beamline 5U.1 at the Daresbury SRS consists of a one metre long, ten period, SmCo₅ permanent magnet with a remnant magnetic field of 0.9T. In common with many undulator devices the magnetic field can be varied by altering the gap between the two planes of magnets. This changes \( K \) and thus the harmonic wavelength, with the minimum gap of 42mm the peak on axis field reaches 0.35T, resulting in a deflection factor, \( K \), of 3.2. With the synchrotron ring electron energy of 2GeV, the fundamental, given by equation 1.5, then occurs at 60 eV, with harmonic radiation beyond 1keV as shown by figure 1.10a.

Of more interest to us was the polarisation response of the undulator as shown in figure 1.10b. It can be seen that the first harmonic (at 60 eV) is 98% polarised whereas the region between the first and second harmonics is only 41% polarised. By varying the undulator gap, and viewing the flux, one can select the fundamental and the region between fundamental and first harmonic for any energy between 60 and ~ 200eV. Thus for such energies polarisation measurements could be made with a 100% and 40% input polarised beam, with all other parameters unchanged. This procedure, described in detail in Chapter 3, allowed our most rigorous test of the X-ray vectorial effect.

The schematic layout of the Beamline is shown in fig 1.11. Photons from the undulator are first reflected by a 250mm platinum coated SiC plane mirror onto a 1200 l/mm grating. The grating is an ion-etched laminar profile plane grating with a platinum coated CVD-SiC surface on a graphite substrate. The focusing element is a 298.8m radius of curvature spherical mirror operated at a grazing angle of incidence of 2°. Operation of the monochromator such that the focal plane of the spherical mirror is independent of energy is possible, allowing, by suitable choice of exit slit, the resolution to be source size limited.
Figure 1.10: a) Calculated flux output from the undulator into a 1.5 x 20 mm (VxH) slit at 10m from source. b) Predicted degree of linear polarisation in the beam from 1.5 x 20mm slit (Roper 1994).
Figure 1.11: Schematic of the optical layout of beamline 5U1 in plan and elevation (Roper 1994)
Chapter 2

Astronomical X-ray Polarimetry

2.1 Introduction

X-ray astronomy remains a relatively young science, with the first extra-solar observation made in 1962 (Giacconi et al. 1962). In the following thirty-five years tremendous progress has been made, both in terms of the numbers (over 50 thousand sources discovered by ROSAT) and types of sources discovered and in the models developed to describe the underlying emission mechanisms. A major limitation, though, has been the lack of independent parameters to discriminate between models of various classes of object. For compact objects such as pulsars, black hole candidates and active galactic nuclei (AGN) analysis of the emission mechanisms has involved only spectral characteristics and time variability. Often two or three significantly different models have been able to explain the observations successfully. For extended objects, imaging has provided positional information to further aid discrimination between models, but obviously for compact objects imaging is unavailable. X-ray polarimetry offers the opportunity of greater discrimination for both compact and extended objects, yielding two new independent parameters - the direction and degree of linear polarisation (Mészáros et al. 1988).

The importance of X-ray polarimetry was first recognised by Dolan (1965), but to date only one positive celestial observation has been made. In 1971 a sounding rocket experiment using both Bragg crystal and Thomson scattering polarimeters measured a linearly polarised fraction
of 15.4 ± 5.2 % for the Crab Nebula at 2.6keV (Novick et al. 1972). A later measurement by a Bragg crystal detector onboard the OSO-8 satellite measured the linearly polarised fraction of the Crab at 2.6keV to be 19% (at the 18 σ level) (Weisskopf et al. 1976; Weisskopf et al. 1978). A marginally significant measurement on Cygnus X-1 of 3% (at 2.6keV) has also been reported (Long, Chanan & Novick 1980). Other measurements were made by Ariel-5 (Griffiths, Ricketts & Cooke 1977; Gowen et al. 1977) and the OSO-8 satellite, which produced, in total, mainly upper limits on 15 sources (Hughes, Long & Novick 1984). Both these satellites employed Bragg crystal polarimeters and these instruments lacked the sensitivity to make definitive measurements on any but the brightest X-ray sources.

A review of the astrophysical production of polarised X-rays and the significance of their measurement to our understanding of various types of source constitutes Section 2.2. Section 2.3 then reviews the type of instrument capable of detecting X-ray polarisation, including those to be flown on the Spectrum X-Gamma satellite. The remainder of the chapter then describes the background to an alternative polarimeter based on the "X-ray vectorial effect".

2.2 Astrophysical Context

Polarised X-rays can be expected whenever emission occurs by a non-thermal process, such as; (1) Synchrotron Radiation, (2) linear bremsstrahlung occurring from mono-directional electrons impinging on an atmosphere, (3) emission from asymmetric hot plasma with densities such that significant scattering occurs and (4) pulsar emission. Measurement of the degree, direction and energy dependence of the linear polarisation provide a measure of the non-thermal electron distribution and magnetic field distributions within the emission region.

Synchrotron (magnetic bremsstrahlung) emission is produced by a flux of relativistic electrons trapped in a quasi-homogeneous magnetic field (Landecker 1972). The trapped electrons spiral around the magnetic field lines generated within sources such as jets, AGN and BL Lac objects, radiating X-rays with the electric vector perpendicular to the magnetic field. Strongly linearly polarised X-rays with the direction of polarisation independent of energy would thus be expected. This production mechanism is generally accepted for the continuum emission from the Crab Nebula as the X-ray polarisation of 19%, is consistent with that at radio and optical wavelengths.
(Novick et al. 1972). Note a pure synchrotron source viewed in the electron plane would produce almost 100% polarised X-rays. That the Crab doesn’t present unity polarisation shows the viewing angle isn’t ideal, some of the flux is not from synchrotron emission or the magnetic fields have large scale inhomogeneities.

Linear bremsstrahlung would produce linearly polarised X-rays as a result of high energy electrons colliding with a cool dense atmosphere. The direction of polarisation would, however, be dependent on the X-ray energy with low energy X-rays being polarised perpendicular to the plane formed by the incident electrons and the resultant X-ray beam. High energy X-rays, on the other hand, would be polarised parallel to this plane. Figure 2.1a shows the variation of polarisation with frequency of the emitted photon for three electron energies (V in eV). The reversal of the state of polarisation which is characteristic of bremsstrahlung (Novick 1975), can be seen to occur between ten and twenty percent of $\nu_0$, where $\nu_0$ is the high frequency cut off and $\hbar\nu_0=E$, the electron’s energy. Figure 2.1a is for the ideal case where the observer is at 90° to the direction of electron motion; figure 2.1b describes the variation of polarisation with this observation angle for electrons with five energy distributions described by $N(E)=N_0 e^{-E/E_0}$. Scattering and X-ray flux arising from electron motion in all directions will significantly reduce the polarisation from the values predicted in these figures.

Thermal bremsstrahlung would only be expected to produce a polarised flux of a few percent, and then only if the isothermal hot plasma was sufficiently asymmetric, and where the density of the electrons is sufficient to produce significant internal Thomson scattering (Angel 1969; Landecker 1972).

Both accreting X-ray pulsars and rotation powered pulsars are expected to produce linearly polarised X-rays. Suggested emission processes include cyclotron radiation from the accreting material, X-rays generated from thermal processes near the stellar surface and synchrotron radiation (Rees 1975; Mészáros et al. 1988). In all cases the X-rays would exhibit strong linear polarisation of up to 80% below 10keV and near the cyclotron resonance energy (normally around 40keV).

Active Galactic Nuclei (AGN) provide a good example of how X-ray polarimetry data would improve our understanding of the physical processes of X-ray emission. AGN emit most of their
Figure 2.1: a) The polarisation of Bremsstrahlung radiation with respect to the frequency of the emitted X-ray for monochromatic electrons of three different energies (V in eV). This is for the case of the observer at 90° to the direction of the incident electron beam. The atomic number, Z, is of order one for an astronomical target. b) The polarisation of bremsstrahlung as a function of the angle between electron direction and the observer for electrons possessing the exponential energy spectrum $N(E) = N_0 e^{-E/E_0}$. $E_0$ values in eV (Dolan 1965).

Radiation in X-rays, but it is unclear if the radiation is thermal or non-thermal, from a disk, torus, quasi spherical flow or a jet. Models based on various combinations of these emission mechanisms can match the spectral and time variation data available. Where the models do differ significantly is in their prediction for the degree of X-ray polarisation. These range from less than a few percent for asymmetrical plasmas, 2-10% for disks to greater than 10-20% for jets. Thus sensitive measurement of the X-ray polarisation would conclusively discriminate between the proposed models (Mészáros et al. 1988).

X-ray polarimetry also has the capability to clearly determine whether an object is a black hole (Rees 1975; Novick, Chanan & Helfand 1985; Mészáros et al. 1988). Current black hole models for objects such as Cygnus X-1 propose that the X-rays emanate from an accretion disk surrounding the central object. The X-ray emission from such a system would be expected to be linearly polarised due to electron scattering in the disk. However, the polarisation angle (the photon electric vector phase angle with respect to astronomical north) is effected by relativistic effects associated with the strong gravitational field extending into the disk. The Lense-Thirring effect (the "dragging of inertial frames") causes a rotation in the plane of polarisation. Furthermore,
2.3 X-ray Polarimeters : General Principles

An ideal polarimeter would have 100% detection efficiency for one linear polarisation state and zero efficiency for the orthogonal state. For many types of polarimeter this is approximated by employing different reflection or transmission properties for photons of differing polarisation. Either way, real polarimeters fall short of the ideal. For any polarimeter a modulation factor $M$ is defined from the count rate modulation for a 100% linearly polarised beam;

![Figure 2.2: The net X-ray polarisation $P(E,\theta)$ from an accretion disk plotted as a function of photon energy for three different black hole masses. $P_c(\theta)$ is the polarisation computed by Chandrasekhar (1960) for a plane-parallel scattering atmosphere and $\theta$ is the incident angle of the disk plane.](image-url)

The rotation increases with increasing photon energy, because higher energy X-rays tend to come from smaller radii and hence stronger gravitational fields. Lightman & Shapiro (1975) modelled the polarisation versus energy function as shown in figure 2.2. Rotation of the plane of X-ray polarisation, coupled with an inferred mass greater than three solar masses would be overwhelming, unambiguous evidence for a black hole.
where $C_\parallel$ and $C_\perp$ are the signal counts when the electric vector is parallel and perpendicular to the most efficient axis of detection. For an ideal polarimeter $M$ is unity.

The determination of source polarisation, $P$ is then by means of the equation:

$$P = \frac{1}{M} \frac{C_{\text{max}} - C_{\text{min}}}{C_{\text{max}} + C_{\text{min}}}$$

where $C_{\text{max}}$ and $C_{\text{min}}$ are the maximum and minimum signal count rates respectively.

The minimum source polarisation which can be detected at the 99% confidence level in an observation time $T$, given by Novick (1974), is:

$$MDP = \frac{3}{MS} \sqrt{\frac{2(S + B)}{T}}$$

This limiting sensitivity is referred to as the "Minimum Detectable Polarisation", where $S$ is the signal count rate and $B$ the background count rate. As a result of photon counting statistics, all measurements, even those made on an unpolarised source, give a non-zero value of $P$. The minimum detectable polarisation is used to quantify the level at which measurements become statistically significant.

A figure of merit, $F$, for comparison of different polarimeter types is, by inspection of equation 2.3 in the strong source limit where $S \gg B$:

$$F = Q^{-\frac{1}{2}} M^{-1}$$

where $Q$ is the detector quantum efficiency. For the ideal polarimeter $F$ equals unity, but for a real polarimeter $F$ would be much greater than unity (Fraser, Lees & Pearson 1989).

Instrumentation for X-ray polarimetry has been extensively reviewed by Novick (1974) where various potential polarimetric mechanisms were evaluated for astronomical use. Gas photoelectric polarimeters, secondary fluorescence polarimeters (Kifune 1972; Weisskopf et al. 1985) and Borrman-effect polarimeters (Cole, Chambers & Wood 1961; Dolan 1966) were all found to be unsuitable for astronomical application. Two other types of instrument, the Bragg crystal
polarimeter and the Thomson scattering polarimeter, were shown to have sufficient sensitivity for X-ray astronomy and are described below. Since then, three additional types of polarimeter have been described, each based on the anisotropy of emission directions of photoelectrons produced by polarised X-rays.

The direction of primary photoelectron emission is preferentially parallel to the electric vector of the incident photon. For emission from a spherically symmetric s shell, the cross section is given by:

\[
\frac{d\sigma}{d\Omega} \propto \sin^2 \phi \cos^2 \phi
\]

where \( \phi \) is the angle between the electron ejection direction and the incident beam and \( \phi \) is the angle between the ejection direction and the electric vector of the photon. Calculation and experiment by Afans'ev et al. (1988) has described a similar, but somewhat less pronounced, anisotropy in the 2p shell.

Detection of the direction of the primary photoelectron is, however, complicated by the fact that the incident X-ray penetrates the detection material to a greater depth than the escape length of the photoelectron. Each of the three techniques described below are, essentially, attempts to get over this problem.

In silicon CCD devices (McLean 1994) the primary charge cloud produced extends parallel to the photon electric vector and if the pixel size is small enough, into neighbouring pixels in that direction, as shown in figure 2.3. Thus, by selecting only events extending into several pixels some of the polarisation information in the incident X-ray beam can be analysed by the CCD. Using 12\( \mu \)m square pixels a small degree of modulation has been detected between 15 and 37 keV (Tsunemi et al. 1992). For a fixed pixel size the polarimetric response improves at higher energies (as photoelectron ranges become longer). But at higher energies (\( E > 40\text{keV} \)) the CCD depletion region (\( d \leq 40\mu\text{m} \)) is insufficient to efficiently stop the X-rays. A significant improvement in performance can thus only arise from using smaller CCD pixel sizes. Holland et al. (1995) modelled modulations of up to 34% using 4\( \mu \)m square pixels, although at this energy (40keV) the quantum efficiency is less than 1%. To date the smallest pixel sizes demonstrated are 4 \( \times \) 9 \( \mu \)m. A CCD polarimeter does, though, have the advantage of simultaneously providing an X-ray image and spectrum as well as polarimetric information.
Figure 2.3: Operating principle of a CCD pixel polarimeter. The anisotropic distribution of photoelectron emission directions (dashed line) causes an excess of events where charge spreads between the central pixel and pixel 1 or 3, which lie aligned to the X-ray electric vector (Holland et al. 1995).

Micro-gap gas proportional counters are a modern incarnation of the gas counter geometries reviewed by Novick (1974). The X-ray is absorbed within a thin (~ 3mm) gas layer. The photoelectron then ionises atoms in its path producing a charge cloud. Anisotropies in the primary charge cloud are detected by an array of closely spaced anode and cathode lines mounted on the base of the detector. Soffitta et al. (1995) using a Ne-DME gas mixture have demonstrated a modulation factor peaking at (11 ± 2)% at 8keV. The major limitation of the device is the gas absorption efficiency which can be moderately improved by increasing the gap thickness to a level where the modulation is not affected by diffusion in the drift. Micro-gap gas proportional counters also have imaging and spectral capabilities.

The third type of photoelectric polarimeter, is based on at least some of the directional information in the primary photoelectrons being passed on to secondary electrons, which then go on to escape the sample. It is this "vectorial effect" that is examined in the following three chapters.
2.3.1 Bragg Crystal Polarimeter

A Bragg crystal operating at an angle of 45° acts as a perfect polarisation analyser over the fraction of an electron volt bandwidth of the Bragg reflection. X-rays which satisfy the Bragg condition and whose electric vector is perpendicular to the plane defined by the incident and reflected rays and the surface normal are preferentially reflected (Bragg 1914). X-rays with electric vectors parallel to this plane are photoelectrically absorbed. A greater bandwidth, which is required to increase the measurable stellar flux, is obtained by using a mosaic of nearly perfect crystals or an ideally-imperfect crystal (Novick 1975).

Theoretical and experimental studies also conducted by Novick (1975) have shown graphite mosaic crystals are the best suited analysers at energies $\geq 1$keV. First order reflection from graphite occurs at 2.6keV, with usable second and third orders at 5.2 and 7.8keV respectively. Natural crystals other than graphite can be used in the energy range of 0.9keV (Mica) to 4.37keV (Lithium Fluoride) with higher orders ($n \geq 2$) above 10keV. Below 1keV the use of artificial crystals is required, these are produced by depositing alternative layers of high and low density materials with periodic spacings of order $10\text{Å}$. In this manner the range 0.1 to 1keV can in principle, be covered.
Figure 2.4 illustrates the design of a Bragg Crystal Polarimeter of the type used on the OSO-8 satellite. A multiwire proportional counter is used as the detector; a focusing mosaic crystal is used to reduce the size of the proportional counter and hence reduce the background noise level \(B\) in eq.2.3, yielding a significant advantage over the plane crystal with large detectors used on Ariel V.

The OSO-8 experiment was optimised for energies above 1keV, the region where near peak intensities for several important classes of X-ray source (e.g. X-ray binaries, pulsars and bursters) lie. Graphite was thus the chosen material. A 200 - 300 \(\mu\)m thick matrix of graphite crystals was mounted on the surface of a paraboloid of revolution. The curvature of the crystal panel encompassed a range of Bragg angles between 40° and 50°, increasing the device bandwidth to 0.4keV for first order reflection and 0.8keV for second, but reducing modulation, in the proportional counter count rate, from unity to 96%.

To function as an analyser, a Bragg crystal is rotated in azimuth about the line of sight of the source. The signal counting rate will be modulated at twice the detector rotation frequency, \(\omega\). The amplitude of the \(2\omega\) modulation component yields the fractional linear polarisation \(P\) while the phase yields the position angle of the electric vector. Note that any instrumentally induced modulation arising mainly from misalignment of the rotation axis with respect to the line of sight should coincide with the rotation frequency and can thus be discriminated against. The result of a Monte-Carlo simulation of the effect of a 0.1° spin axis misalignment on the Stellar X-Ray Polarimeter (SXRP) Bragg crystal polarimeter is shown in figure 2.5. The \(2\omega\) modulation component is 96% ± 0.3%, but \(1\omega\) and \(3\omega\) modulations of 24% are present for the 0.1° misalignment case.

### 2.3.2 Thomson Scattering Polarimeter

The Thomson scattering polarimeter is based on the principle that X-rays are preferentially scattered orthogonally to the incident photon electric field vector (Landecker 1972). The angular dependence of the scattering cross section, as illustrated by figure 2.6a, is proportional to \(\sin^2\phi\), where \(\phi\) is the angle between the polarisation vector of the incident photon and the propagation vector of the scattered X-ray.
Figure 2.5: Monte Carlo simulation of the effect of a 0.1° misalignment in the rotation axis of the Bragg crystal polarimeter (marked 0.1) and for perfect alignment (0.0) (Silver et al. 1994).

Figure 2.6b shows the schematic design of a Thomson scattering polarimeter to be used in SXRP, where the detectors surrounding the scattering block are proportional counters. Ideally the scattering block would be entirely encased by a cylindrical "wraparound" proportional counter. The polarimeter could in principle operate without rotation and the modulation derived from the different count rates in orthogonally situated detectors. In practice, though, the instrument is always rotated about the line of sight to the source so that any differences in efficiency between the detectors are not interpreted as source polarisation. The modulation factor for the Thomson scattering polarimeter on SXRP has been modelled as peaking at 77% at 6 keV (Elsner 1990). The modulation factor could be improved further by reducing the solid angle subtended by the proportional counters, but only at the cost of reduced efficiency.

The energy range of a Thomson polarimeter is generally 4 - 10 keV. The range is limited at the lower end by photoelectric absorption within the block while the higher end is limited by decreasing absorption in the proportional counter, a decrease in flux from celestial sources and a fall off in mirror throughput. A low-Z material is generally used to enhance the scattering-to-absorption ratio, with metallic lithium (Novick, Chanan & Helfand 1985), LiH (Landecker 1972) or beryllium (Mészáros et al. 1988; Weiskopf et al. 1994) being common choices. In the
case of a lithium block, encapsulation in a thin beryllium container is required to limit oxidation effects. A Thomson polarimeter, unlike the narrow band Bragg crystal polarimeter, can measure polarisation over a broad energy range.

2.3.3 The Stellar X-ray Polarimeter (SXRP) Instrument on Spectrum X-Gamma

An advantage of Bragg crystal and Thomson scattering polarimeters is their ability to be used in conjunction with one another. If the Bragg crystal is sufficiently thin, photons can first meet the Bragg crystal where a very small percentage are reflected to a proportional counter, with the vast majority passing straight through and becoming available to the Thomson scattering polarimeter. This arrangement is to be utilised on the only X-ray polarimeter to be launched in the next few years. The Stellar X-Ray Polarimeter (SXRP) (Silver et al. 1994) is one of the many instruments on the Russian/W. European/USA Spectrum X-Gamma mission scheduled for launch in 1998. Four imaging proportional counters (IPC) surround the two polarisation analysers, with one IPC detecting photons from both the Thomson scatterer and the Bragg crystal (Kaaret et al. 1991).

A flat graphite Bragg polarimeter is tilted at 45° to the optical axis. The graphite is a mosaic of crystals with random misalignments of order 1°. The spread of angles in the converging beam produce a device bandwidth, \( \Delta E/E \) of \( \sim 0.08 \). The scattering target is a cylinder of metallic lithium, 3cm in diameter and 7cm long housed in a 125\( \mu \)m beryllium container to protect it from atmospheric contamination. Monte Carlo model predictions for the effective area of the Thomson Scattering peaks at 65cm\(^2\) (c.f. mirror area of 1200cm\(^2\)) at 9keV with a modulation factor of between 0.71 and 0.77 (Elsner 1990). The predicted Minimum Detectable Polarisation (Equation 2.3) for a 10\(^8\) second observation using both polarimeters simultaneously is then given in Figure 2.7.

SXRP will be an order of magnitude more sensitive to polarisation than previous satellite experiments. Most of this improvement is due to the fact that SXRP sits at the focus of a large x-ray optic, the SODART mirror assembly (effective area \( \sim 1200\)cm\(^2\) at 7keV (Kaaret et al. 1993)). This is also a disadvantage as SXRP must share the focus with an imaging detector and
Figure 2.6: (a) The Angular Variation of X-ray Scattering (Landecker 1972), (b) Thomson Scattering Polarimeter schematic
Figure 2.7: Minimum detectable polarisation of the SXRP graphite and lithium polarimeters as a function of source strength for a Crab-like spectrum. The three curves are for first order Bragg reflection at 2.6 keV, for second order Bragg reflection at 5.2 keV and for the lithium Thomson polarimeter over the energy band 6 to 12 keV. A nett background flux of 0.003 counts/cm² sec keV was assumed (Kaaret et al. 1993). A bright AGN has a flux between 1 and 10 mCrab.

A spectrometer. SXRP is thus only guaranteed 11 days observation time per year. With that time SXRP is expected to increase the number of positive polarimetric measurements from one to a few dozen, but measurements on many classes of fainter, yet interesting sources such as AGN will still not be possible.

2.3.4 Comparison of X-ray polarimeter designs

A comparison of polarimeter designs at a given energy is possible using the figure of merit as defined by equation 2.4. Astronomical sources are broadband, so the bandwidth of the detector also needs to be accounted for. A fairer figure of merit comparison is thus;

\[ F' = Q^{-\frac{1}{2}} \delta E^{-\frac{1}{2}} M^{-1} \]  

(2.6)
detectors with higher quantum efficiencies and similarly high modulation factors can be developed, or an efficient polarimeter can be developed in the lower energy X-ray range (0.05 - 1 keV) where source strength is far higher, although the degree of polarisation for various classes of source object is expected to be lower.

The X-ray vectorial effect offers the possibility of both the above benefits, with the opportunity to use a highly efficient caesium iodide photocathode detector over a bandpass from 0.1 to 3keV.

2.4.1 Theoretical Considerations

X-rays impinging on a solid material have the energy to free electrons from atoms within the solid; some of these primary electrons are also of high enough energy to release further electrons from neighbouring atoms. In alkali halides such as Caesium Iodide an X-ray of a few keV energy, can release up to 10 electrons into vacuum from the solid photocathode (Eliseenko, Shchemelev & Runah 1968). An idealised energy spectrum of emitted electrons excited by X-rays is shown in figure 2.8. The energy spectrum consists of primary (high energy) photoelectrons, Auger electrons and a large number of low energy electrons. These low energy electrons, with an energy of under 50eV, are known as secondary electrons and typically dominate the total photoyield. In CsI this is taken to the extreme with secondary electrons constituting over 99% of the total for a normal incidence X-ray (Henke, Knauer & Premaratne 1981). It should be noted that the energy distribution of secondary electrons depends on the angle of incidence, with higher energy secondaries being produced at the grazing angles of interest for the vectorial effect (Ausmees et al. 1990).

In this situation two polarisation states, S and P are defined thus; in the P state the electric vector of the X-ray is parallel to the plane of incidence (defined by the photon trajectory and the surface normal) whereas in the S polarisation state, the electric vector is perpendicular to the plane of incidence. The emission of a photoelectron is known to be preferentially in a direction parallel to that of the X-ray's electric vector. For X-ray absorption in a spherically symmetric s-shell the angular distribution is described by equation 2.5. From an asymmetric shell the distribution is less pronounced, but still favours emission parallel to the electric vector (Afans'ev et al. 1988). Other than at normal incidence, the electric vector has a component out
Figure 2.8: Example energy spectrum of photoelectrons emitted from a solid photocathode under X-ray bombardment, the y axis representing number of such electrons. The secondary electrons, which typically form the largest fraction of emitted electrons, form a narrow distribution below 10eV. The higher energy primary electrons consist of elastically scattered photoelectron and Auger electron sharp "lines" and a continuum of inelastically scattered electrons in their low-energy tail. (Henke, Knauer & Premaratne 1981)

of the surface for p-polarised photons and in the surface for s-polarised light. The basis for the x-ray vectorial effect is that some of the polarisation sensitivity of the primary photoelectrons would be transported to the secondaries. It is also possible that the optical properties of the surface are significantly different for the two polarisation states.

2.4.2 Previous Observations

In 1894 optical photoyield was found to be dependent on the linear polarisation state of the incident light, when it was shown that the ratio of p to s photocurrents from a liquid Na/K alloy could greatly exceed both unity and the corresponding ratio of absorbed intensities (Elster & Geitel 1894). The first UV measurements were conducted by researchers at the University of Southern California who found the photoemission of Gold and Aluminium was polarisation dependent (Morse 1967). A group at the University of Windsor, Canada later described a polarisation term in the quantum efficiencies of lead oxide microchannel plates (McConkey, Crouch & Tomc 1982; Tomc et al. 1984) and in the photoemission from planar Au and PbO photocathodes (Zetner et al. 1983; Zetner et al. 1984). The polar diagram of figure 2.9 shows
the data of McConkey, Crouch & Tomc (1982) for the efficiency of an MCP detector at various angles of azimuth.

The first X-ray measurements were conducted at the Photon Factory by the Hamamatsu Corporation on CsI, Al₂O₃ and PbO. X-rays from a synchrotron source were shone onto each sample at a range of energies and angles of incidence; electrons emitted from the photocathodes being collected by a separate detector. Each photocathode material displayed non zero modulations, of increasing magnitude toward grazing incidence. As is demonstrated by figure 2.10, even CsI showed pronounced polarisation sensitivity even though emission is virtually all secondary electrons. The possibility that such a highly efficient photocathode could form the basis of a practical polarimeter led to the construction of a custom built polarimeter at Leicester in 1988 (Lees 1989).

Figure 2.11 shows a schematic of the Leicester photoemission polarimeter. X-rays are produced when electrons from a filament are accelerated to the anode; the X-rays are then collimated and polarised by right angle Bragg reflection. Four different Bragg crystals are used to provide a choice of four X-ray energies between 0.175 and 2.7 keV. Each of the crystals was also found to specularly reflect UV radiation. Two filters were thus made available in the vacuum system; a polypropylene filter could be used to block the UV component, or a sapphire window could be inserted to allow only the UV component through to the photocathode. Using neither filter produced a broad band UV beam with a single X-ray line energy, with the vast majority of the flux in the UV. Polarisated photons of the desired energy then eject electrons from the photocathode material, the electrons are funnelled by the potential of the mesh cone to the Channel Electron Multiplier (CEM) and pulse counting electronics. By precise alignment of the rotational axis of the photocathode to the optical axis, two important sets of polarisation sensitivity measurements could be made.

i Measurement of the variation in CEM count rate as the photocathode was rotated through 360° in azimuth about the optical axis with constant angle of incidence θ (known as a φ scan)

ii Measurement of the count rate at each of two orthogonal φ settings while varying the angle of incidence ("θ scans")
Figure 2.9: The polarisation sensitivity of an MCP for normal incident photons. The radial distance to the data points (os) represents the counts for the respective azimuthal angles and the central arrow indicates the direction of residual polarisation from the VUV ($\lambda \sim 100$nm) source. The modulation factor by equation 2.1 is 0.125 (McConkey, Crouch & Tomc 1982).

Figure 2.10: Relative sensitivities of a CsI photocathode to 7.5 nm (320eV) S- and P-polarised radiation at a range of angles of incidence (K. Oba, private communication).
Figure 2.12 shows typical results for both types of measurement. (a) is a UV φ scan demonstrating the 180° modulation characteristic of polarisation. Peaks in the photoyield variation at azimuthal angles of 0° and 180° correspond to a P polarised photon beam, while 90° and 270° correspond to S polarisation. The UV band pass is taken to be defined by the sapphire filter as being between 1300 and 5000 Å. Figure 2.12b is a θ scan at 1.65 keV detailing a reversal of most sensitive polarisation state (i.e. change of sign of M) at ~15° grazing angle. More detailed accounts of these experiments (Lees 1989; Fraser, Lees & Pearson 1989; Fraser et al. 1989; Fraser, Lees & Pearson 1990) stress the importance of measures to avoid spurious instrumental polarisation. The problems associated with the aligning the axis of photocathode azimuthal rotation to the polarised photon beam in a system of limited signal to noise is compounded by the problem of ensuring uniform electron collection over the large photocathode illuminated area at grazing incidence. In 1990, a new polarisation system was built to minimise these problems. A schematic would look very similar to figure 2.11, but the apparatus arms (source to crystal and crystal to detector distances) were lengthened to produce something closer to a parallel beam. A focusing Bragg crystal was also installed in place of the flat one (in figure 2.11) to produce a spot on the photocathode and remove the inhomogeneities of electron collection. Since that time problems with the movement of the detector assembly under gravity as φ was altered, a low X-ray flux and the availability of synchrotron measurements have led to a decline in use of the laboratory system.

The respectable modulations obtained at the various X-ray line energies led the Leicester group to commence similar experiments at the Daresbury Synchrotron Radiation Source (SRS) in June 1990. The highly polarised synchrotron beam coupled with X-ray energy tunability over a broad band allowed more detailed analysis of the polarisation signature of CsI and other photocathode materials.

Beamline 6.1 (Section 1.3.1) of the Daresbury SRS was selected for its energy range of 40 - 200 eV. Due to the nature of the experimental chamber of this beamline φ scans were not possible; instead θ and φ were fixed and the photon energy varied throughout the available range. The photocathode was then rotated 90° in azimuth (with fixed grazing angle θ) and the energy scan repeated for the other polarisation state. Measurements on CsI photocathodes were compromised by significant radiation damage (Chapters 8 and 9) although some polarisation sensitivity was still visible (Fraser et al. 1991a). Figure 2.13 shows similar measurements made
Figure 2.12: a) UV $\phi$ scan of a CsI coated photocathode at constant grazing angle of incidence $23^\circ$ (Fraser, Lees & Pearson 1990). b) $\theta$ scan of a CsI photocathode under 1.647keV X-ray illumination. Filled circles denote p-polarisation ($\phi = -90^\circ$); open circles s-polarisation ($\phi = 0^\circ$); crosses represent the relative photoyield for unpolarised radiation (i.e. the average of s and p polarisation states) (Fraser, Lees & Pearson 1989).
on a gold photocathode, where apparent modulations of up to 40% were measured for a 10° grazing angle between 60 and 200 eV. Note that over this range there are four atomic absorption edges (dotted vertical lines on the figure) which have no effect on the observed modulations. The figure also shows measurements taken 10 hours apart to show the absence of radiation damage effects in metallic cathodes.

The P state can be seen to be dominant, thus producing by equation 2.1, a negative modulation factor. This is as expected from equation 2.5 which has the primary photoelectron preferentially ejected along the line of the electric vector, which for grazing incidence measurements, is in the plane of the surface for S state and out of it for P state. P state dominance is also expected as a result of differences in Fresnel reflectivity coefficients between the polarisation states.

Quantum efficiency versus angle of incidence measurements (θ scans) were also completed to aid the understanding of the physical mechanisms behind the modulation signatures. Figure 2.14a illustrates the photoyield dependence on grazing angle, the important point being the yield curves for the two polarisation states have different shapes. This would tend to discount angular misalignment as the cause of the modulation. Misalignment, whereby the grazing angle of incidence was different for each polarisation state is the most likely cause of spurious modulations, but would lead to θ scans displaced along the θ axis, not as seen in figure 2.14a. Figure 2.14b shows the dependence of the modulation factor on the grazing angle of incidence.

The solid line in figure 2.14 is the calculated modulation caused by differences in the Fresnel reflectivity coefficients for orthogonal linear polarisation states. For a uniaxial material with complex refractive indices, η, in the transverse and longitudinal directions given by:

\[ η_1 = 1 - δ_1 - iβ_1 \]
\[ η_2 = 1 - δ_2 - iβ_2 \] (2.7)

Longitudinal and transverse dielectric constants, ɛ, are then given by:

\[ ɛ_1 = 1 - α_1 - iγ_1 \]
\[ ɛ_2 = 1 - α_2 - iγ_2 \] (2.8)

where \( α_n = 2δ_n - δ_n^2 + β_n^2 \) and \( γ_n = 2(1 - δ_n)β_n \).
Figure 2.13: Quantum efficiency of a gold photocathode for s and p polarised X-rays at 10° grazing incidence. The modulation calculated (Eqn 2.1) from these efficiencies is shown in the lower diagram. The vertical dotted lines represent the four gold absorption edges in this energy range. Both figures contain a second data set taking 10 hours later to demonstrate the reproducibility of the results (Fraser et al. 1991b)
The reflectivity for s polarised light at the interface between vacuum and a uniaxial photocathode of thickness greater than the electron escape depth is given by Pepper (1970) as:

\[ R_s(\theta) = \left| \frac{\sin \theta - (\epsilon_i - \cos^2 \theta)^{1/2}}{\sin \theta + (\epsilon_i - \cos^2 \theta)^{1/2}} \right|^2 \]  
(2.9)

and similarly for p polarised light:

\[ R_p(\theta) = \left| \frac{\epsilon_i \sin \theta - (\epsilon_i)^{1/2} (\epsilon_i - \cos^2 \theta)^{1/2}}{\epsilon_i \sin \theta + (\epsilon_i)^{1/2} (\epsilon_i - \cos^2 \theta)^{1/2}} \right|^2 \]  
(2.10)

In the isotropic limit (\( \epsilon_i = \epsilon_l \)) these reflectivities reduce to the familiar forms given by Henke (1972) while at normal incidence and irrespective of the ratio \( \frac{\epsilon_i}{\epsilon_l} \), \( R_s(90^\circ) = R_p(90^\circ) \) as expected.

The modulation due to differences in the reflectivities to photons of orthogonal polarisation state is then:

\[ M(\theta) = \frac{R_p - R_s}{2 - R_p - R_s} \]  
(2.11)

The modulation measured here (filled circles) cannot be ascribed to reflection induced modulation, the full curve in figure 2.14b. Corresponding measurements on aluminium and Al\(_2\)O\(_3\) also showed large apparent modulations, although with significantly different energy and angular profiles.

Beginning in 1988, a group at the Columbia Astrophysical Laboratory (CAL) became interested in the vectorial effect and designed their own photoemission polarimeter (Heckler et al. (1989), Shaw et al. (1990), Figure 2.15). The experimental configuration was similar to that of the Leicester system described above. The major difference was the use in the CAL system of a Microchannel Plate (MCP) detector for the electrons emitted from the photocathode. This detector operated in what is known as a pulse counting mode, that is it has essentially no resolution of the number of electrons reaching the MCP, whereas the CEM measures the number of electrons emitted. Initial CAL measurements at 2.69keV detected similar modulation profiles, however after significant refinement to eliminate spurious causes of polarisation sensitivity, a more accurate experiment measured a modulation factor of effectively zero (Hanany et al. 1993). CAL also found that earlier experimental inaccuracies could fully explain their previously measured modulation curves without requiring a physical vectorial effect (Shaw et al. 1991). The CAL group also conducted an experiment at the National Synchrotron Light Source (NSLS) at Brookhaven.
Figure 2.14: a) Variation of 115eV quantum efficiency with grazing angle for gold. Circles - Fraser, Lees & Pearson (1991) data at p and s polarisations. Full curve - estimate of $\chi_c$ for unpolarised radiation (average of s and p yields). Broken curve - unpolarised pulse quantum yield data of (Savinov & Lukirskii 1967). b) Comparison of measured and calculated ("Fresnel") modulation factors $M(\theta)$ for a gold photocathode at 102eV.
Figure 2.15: The Columbia Astrophysical Laboratory (CAL) laboratory polarimeter. The electron detector is a pulse counting microchannel plate fronted by two potential grids to define the energy of detected electrons (Hanany et al. 1993).

using a gold photocathode. A highly collimated beam of monochromatic X-rays (of energy 2.5, 2.7 or 4keV) reached the photocathode at a fixed grazing angle between 5° and 50°. The azimuthal angle was then rotated between 0° and 360°, the resulting measurements constrained the modulation factor to less than 4% (Shaw et al. 1994).

Obviously it is far easier to detect spurious modulation, than miss an effect which truly is there. Alternatively, both results could be correct with the apparent discrepancy due to the fact different properties are being measured. The Columbia group effectively measured the number of pulses of electrons produced under X-ray illumination, whereas Leicester counted the number of electrons emitted. If the polarisation sensitivity were in the number of electrons per pulse, but not the number of pulses the results could be reconciled. Efficient alkali halide photocathodes can produce up to 20 electrons per photon, at X-ray energies of order a few keV and grazing incidence. Modulations of 10-20% measured at Leicester correspond to difference in efficiency of about 4 electrons per pulse between the S and P polarisation state. The CAL experiment would not be sensitive to the difference between 16 and 20 electrons per pulse and thus see the zero modulation recorded. Such reasoning, however, could not explain the differences in modulation
factor measurements on gold photocathodes where the number of electrons per pulse is nearer unity.

Finally the CAL group conducted an experiment to measure the polarisation sensitivity of their equipment to primary electrons. Using the setup as in figure 2.15 with three meshes in front of the MCP detectors to form a retarding field. A silicon (100) crystal was used as a photocathode for K shell excitation by the 2.69keV X-ray source. Three hundred and sixty degree scans in azimuth were conducted at a range of angles of incidence with a fixed retarding voltage of 100V (to discriminate against secondaries). A modulation factor of 8.4 % at 5° grazing incidence was measured for primary electrons consisting of no-loss photoelectrons and Auger electrons. Measurements were also conducted at fixed azimuthal angles, varying the repelling field. These measurements showed the polarisation sensitivity was present in photoelectrons, but not Auger electrons; when the repelling voltage was large enough to block Augers a modulation of 40% in the remaining signal was recorded (Shaw et al. 1994). It should be noted though that the number of such high energy electrons created was very low. The significance of these measurements to astronomical X-ray polarimetry is discussed in Chapter 5.
Chapter 3

Polarimetric Measurements in the Energy Range 2-6 keV

3.1 Experimental Details

Following the measurements at energies between 60 and 200 eV, it was decided to conduct an experiment at higher energies. Beamline 6.3 was chosen as it allowed us access to 2.69 keV, the energy used for the Columbia Astrophysics Laboratory experiments. Beamline 6.3 is described in section 1.3.2 and in more detail in the beamline reference manual (MacDowell, Norman & West 1986).

A 0.9 mm diameter collimating hole inside the experimental chamber was required to reduce the beam width (and to a lesser extent height). The default spot size at the sample, as detailed in section 1.3.2, is approximately 4 mm wide by 1 mm high (fwhm) which, given that the investigation of polarisation dependence requires the use of small grazing incidence angles, would produce a very large spot on the photocathode. Too large a spot size would invite the possibility that at certain angles some of the flux would miss the sample completely, compromising the comparability of S and P efficiency measurements. The collimating hole has an energy dependent transmittance, as the position in space of the peak flux changes with energy as the monochromator crystals are rotated (and thus the beam position moves). This problem was solved by
providing another drain current beam monitor (0.75 μm Al foil) behind the collimating aperture. The collimating hole was attached to a linear drive to allow it to be moved across the beam.

Measurements were to be conducted on gold and caesium iodide coated samples. Gold samples were produced at Leicester by evaporating 500Å layers onto stainless steel substrates. The samples were cleaned in situ by argon sputtering (600eV ions for 10 minutes) before commencing measurements at Daresbury. Caesium Iodide samples were prepared at Daresbury to reduce handling. For these evaporation a preparation chamber was connected to the primary experimental chamber, separated by a gate valve. CsI was thermally deposited onto stainless steel substrates at a rate of order 20Ås⁻¹ under a chamber pressure of approximately 5x10⁻⁹ mbar. A 200mm extension pipe was fitted to the evaporator to increase the source-to-substrate distance and ensure even coverage over the 15mm by 15mm active area. A quartz crystal film thickness monitor was to be used to measure the photocathode thickness, however the sample plate totally shadowed the crystal. Thus the sample was moved from in front of the crystal so the deposition rate could be measured and the thicknesses estimated from the rate multiplied by the time. By this method a photocathode thickness of approximately 4800 Å was measured. Samples were then transferred under vacuum to the manipulator arm in the main experimental chamber.

Figure 3.1 shows a simplification of the configuration inside the experimental vacuum chamber at the end of beamline 6.3. The photocathode samples could be rotated in two planes to allow grazing incidence measurements to be made for both polarisation states. As shown 180° rotation in one plane was possible, so two measurements of S polarisation state photoyields were available. This provided a further check on the correct alignment of the rotation axis to the beam axis. If the alignment were imperfect the angles of incidence for S and S' would differ, and thus, so would the emission efficiencies which have a strong angular dependency at kilovolt X-ray energies. Freedom of movement for P polarisation state measurements was more limited with only 120° rotation being available. The 30° rotation into a P' state only allowing a P - P' correspondence check for grazing angles greater than 60°. Besides this rotational freedom, the samples could also be laterally adjusted in x, y and z directions.

Initially a silicon wafer was installed onto the manipulator. This was then used as a reflector for an autocollimator mounted on port 1 (Figure 3.2). The two angular drives, denoted φ (for rotation about a horizontal plane) and α (for rotation about the vertical plane), were then
Figure 3.1: Simplified experimental chamber arrangement showing the sample's freedom of movement.

adjusted until the sample surface was normal to the autocollimator. The procedure was then repeated for port 2; knowing both ports lie on the same vertical plane as the beam defines the normal incidence $\phi$ angle. From engineering drawings of the chamber ports 1 and 2 should lie 80.0° apart with the beam entry mid way between them. The alignment measurements suggested this angle was in fact 80.3° and the beam was assumed to lie mid way between them. Thus both angular positions were accurate to less than 0.3°.

A Gadox phosphor-coated sample was included in the chamber, allowing us to see the position of the beam. The edges of the sample holder and collimating hole were also phosphor coated so the beam should be visible as it moved with rotation of the monochromator crystals.

Due to the physical movement of the beam position with energy, the overall energy range of the beamline was split into three sections (2-3 keV, 3-4 keV and 4-9 keV) with the collimating hole positioned for the centre of each range (or 5keV for the 4-9keV range). Even using this split, the data collected above 6.5keV was of limited quality due to a loss of flux.
The measurement technique following alignment procedures was:

1. Select energy in centre of required range
2. Move collimator hole and beam monitor to maximise current from the beam monitor ($I_0$).
3. Select required angles, move monochromator energy to start of range.
4. With angles fixed, proceed through energy range in steps of order 10eV, recording drain current readings from sample, reference ($I_0$) counter and various beamline monitors.
5. Repeat steps 3 and 4 for each polarisation state for the range of angles of interest.
6. Repeat procedure for next energy range.

### 3.2 Gold Photocathodes

Although gold has a considerably lower quantum efficiency than CsI it is nevertheless a more useful material to examine as it does not suffer the structural degradation that CsI does under intense synchrotron X-ray illumination. A wealth of data was generated over both the lower energy ranges, at grazing angles of incidence of 5, 8, 10, 12, 15, 20, 30, 45, 60 and 90° for S and P
polarisation states. Early in the experimental campaign problems in the data were detected, most importantly the drain current readings for S and S' measurements did not agree. By definition these photocurrents should be identical so it was decided to recalibrate the angular scales. The second calibration was conducted by taking a set of S measurements and then finding the position where S' measurements gave an equal drain current reading. The normal incident point then, by definition, mid way between the two angular settings. A similar method was used to check that the equivalent angle for P was correctly calibrated, although for the less accurate grazing angle of 60°. By this method it was discovered both normal incidence positions were significantly out, the S state (ϕ angle) by 1.0° and the P state (α angle) by 0.65°. Data collected prior to the discovery of the misalignment could be corrected by assuming the efficiency scaled as cosec(θ) where θ is the grazing angle of incidence. Thus, for S and P scans supposedly conducted at angle θ the true efficiencies are:

\[ \chi(S, \theta) = \chi(S, \theta + 1°) \frac{\sin(\theta + 1°)}{\sin\theta} \]  
(3.1)

and

\[ \chi(P, \theta) = \chi(P, \theta + 0.65°) \frac{\sin(\theta + 0.65°)}{\sin\theta} \]  
(3.2)

Figure 3.3 shows the effect of this correction on data taken at 12° grazing incidence. All subsequent data presented in this chapter has either been corrected in this manner or were collected after the misalignment was discovered.

Another problem with the data was that the gold quantum efficiencies recorded for both polarisation states were up to 100% higher than those measured in an earlier Daresbury campaign (Fraser et al. 1992). Figure 3.4 compares the earlier data with that presented here for normal incidence X-rays. The efficiency anomaly was found to be best explained by cross coupling between the chamber reference counter and the sample. Electrons released by the sample are not collected, rather the sample is connected to Earth and the current from Earth to maintain the electrical neutrality of the sample is measured. This same technique is also used for the thin aluminium foil reference counter. If electrons from the sample were to reach the reference counter, this would reduce its drain current reading and thus make us believe less photons were entering the chamber than was really the case. If one assumed the true efficiencies were as measured orig-
Figure 3.3: Quantum efficiencies of gold photocathode to monochromatic S-(+s), S'-(□s) and P- (⊗s) polarised X-rays. The top figure is uncorrected for angular miscalibration creating the large difference in S and S' values. The lower figure is corrected by the cosecθ angle of incidence dependence showing agreement within experimental error. The dashed vertical lines represent two of the M absorption edges of gold as labelled.
Figure 3.4: Unpolarised quantum efficiency of a gold photocathode. □s represent data in the low energy range, xs represent data in the mid energy range and +s previously taken data over a similar energy range (Fraser et al. 1992). Note the almost factor of two difference in measured quantum efficiency and the discrepancy in the overlap region for the two current data sets (□s and xs).

Finally (Fraser et al. 1992) and by discrete line measurements of others (Eliseenko, Shchemelev & Rumsh 1968; Henke, Knauer & Premaratne 1981; Day et al. 1981) it would require a maximum of 12% of the electrons released by the photocathode to reach the reference counter when both were parallel, dropping to less than 1% as the photocathode was rotated to 10° grazing incidence. Qualitatively this mechanism makes sense but no quantitative correction to the efficiency data is possible. This cross coupling should though have no effect on the modulation factors as these are calculated as ratios and, unless any electric field is highly inhomogeneous, the same fraction of electrons should reach the reference counter for each polarisation state. One other complication in the data is visible in figure 3.4, this is the increase in efficiency toward the limits of each data set. This was due to the amount of flux passing through the collimating hole and onto the photocathode decreasing due to beam movement. As the flux decreases an offset on the reference counter readings gained increasing significance, but by comparing a number of overlapping data sets the magnitude of the offset proved easy to calculate and thus correct for.
With the correction for the reference counter offset and, where applicable, the angular correction, the data forms a pleasingly consistent set. Figure 3.5 shows the modulation factor, as defined by equation 2.1, versus energy between 2.1 and 4keV. The modulations are negative, showing P state dominance as seen previously and expected (Fraser et al. 1992). At 8° grazing incidence a 13 % modulation factor can be seen throughout the energy range. An angular dependence is also clearly visible with greater (more negative) modulations for smaller grazing angles of incidence. This is as expected as for θ = 90° the modulation is by definition zero. In fact, figure 3.6 shows the modulation drops below a couple of percent at angles greater than 30°.

The data presented in figure 3.5 is of a high standard, especially given that the gap between high and low energy runs at a particular angle is of order 36 hours, in which time at least one beam refill and repeated repositioning of the sample had occurred. However, the higher grazing angles (Figure 3.6) show that the experimental accuracy is limited, producing greater errors as S and P efficiencies converge.

Figures 3.7, 3.8 and 3.9 display the unpolarised efficiency data for a gold photocathode between 8 and 12°, 15 and 30° and 45 to 90° respectively. These are the mean of the efficiencies to S and P polarisation states from which the modulations are calculated. As stated earlier, these should be considered relative due to the effect of cross coupling.

The existence of a number of required corrections and especially of angle of incidence dependent errors, illustrate the possibility of obtaining a modulation "result" solely due to experimental inaccuracies. Thus a more definitive test was conducted using what became known as gold "angled plates". The idea behind the angled plate is that it can be rotated through S, P and intervening states with the angle of incidence always the same. The plate, as shown in figure 3.10, consisted of a stainless steel body, with a gold coated surface at 10° to the rotational drive shaft. The angled plate was then mounted on a rotational drive and centred by moving the plate though the beam in x, y and z planes separately and recording drain current. A 5° gold angled plate as well as two phosphor coated plates for alignment were also produced but could not be transferred from the storage carousel to the manipulator arm inside the vacuum chamber. The available azimuthal movement for the angled plate was only between -22° and 83° but this would be sufficient to include a pure S state (-7.3°), a pure P state (at 82.7°) and any number of intervening states.
Figure 3.5: Modulation Factors for Gold at 8° (o and +), 10° (△s and xs) and 12° (+s and □s) degrees grazing incidence.

Figure 3.6: Modulation Factors for Gold at 15° (os and xs), 20° (+s and +s) and 30° (△s and □s) degrees grazing incidence. Note that in the higher energy range 20° has a lower modulation than 30°—most likely indicative of the experimental errors.
Figure 3.7: Quantum Efficiency of a gold photocathode to simulated unpolarised X-rays at 8, 10 and 12 degrees grazing incidence
Figure 3.8: Quantum Efficiency of a gold photocathode to X-rays at 15° (os and +s), 20° (Δs and xs) and 30° (*s and □s) grazing incidence.

Figure 3.9: Quantum Efficiency of a gold photocathode to X-rays at 45° (□s and *s), 60° (+s and Δs) grazing incidence and normal incidence (xs and os).
Figure 3.10: A 10° grazing incidence gold coated angled plate.

Measurements were conducted at 2.5keV, 2.7keV and, as shown in figure 3.11, 3.5keV. Unfortunately, at some point in the experiment the various recorded signals were placed in the wrong channels and we ended up watching the variation of counts from the reference counter instead of sample drain current. Coupled with the loss of the phosphor coated plate we failed to notice what is abundantly clear from figure 3.11; the beam was missing the sample at various azimuthal angles.

3.3 Caesium Iodide Photocathodes

Caesium Iodide is probably the best soft X-ray photocathode material due to its high quantum efficiency (Breskin 1996). Thus, for any astronomical polarimeter CsI would be the first choice material. It does, however, suffer degradation of quantum efficiency under synchrotron fluxes, thus complicating experiments to measure the modulation factor (see chapters 8 and 9). To ensure compatibility between measurements for both S and P polarisation states the photocathode surface used for S and P measurements must be fresh or already exposed to an equal X-ray dose. The position of two different spots on the photocathode were defined as shown by figure 3.12. The bombarded areas for S and P measurements can be seen to only just overlap at 8°, the lowest grazing angle used. Measurements of the polarisation response would then be conducted in order S, P, S and P again. This allowed equal recovery times between exposure for S and P measurements and thus hopefully compatible data.

Figure 3.13 shows the derived modulations from S and P polarisation state efficiency measurement at the recorded angles. In contrast to the gold measurements, these data show no agreement between energy ranges or believable angular dependence of modulation. The fact
Figure 3.11: Variation of quantum efficiency with azimuthal angle of incidence measured with the 10° grazing incidence angled plate.

Figure 3.12: S and P bombarded areas during normal energy scans. The extent of the spots is indicated for 5, 6, 8 and 10 degree grazing angles.
Figure 3.13: Modulation Factor measurement on a CsI photocathode between 2 and 4 keV. × and ○) 10° grazing incidence, □s and △s) 12 degrees grazing incidence, +s) normal incidence.

that at normal incidence a 10% modulation is recorded show the experimental errors to be at least of this order as by definition modulation must be zero at normal incidence.

The efficiency data, from which the above figure is generated, is presented in chapter 7 (i.e. figures 7.1 and 7.3) and is relatively consistent in angular dependence and across the energy range. By the nature of modulation measurement only a small error in efficiency measurement is required to produce a "false" modulation of order 10% which, in this case, dominates any true modulation present. As demonstrated by the experiments on radiation damage of alkali halides presented in chapters 8 and 9 a small difference in time the spot had been irradiated could easily cause an error in modulation of this order. An X-ray source significantly less powerful than the synchrotron at full ring current is thus required to test the polarisation sensitivity of CsI photocathodes which from this experiment can only be said to have placed an upper limit on any modulation at ~ 20% in the 2-3 keV band and < 10% in the 3-4 keV band.
3.4 Discussion

Although there were problems in the planar gold photocathode data, it would appear these are well understood and where possible have been corrected for. Cross coupling of electron from the sample reaching the reference counter can explain why the absolute quantum efficiency of the photocathode differs from earlier measurements (i.e. Fraser et al. (1992)). It has been assumed above that cross coupling does not affect the modulation values, an assumption which appears valid for two reasons. Firstly, when the number of electrons required to cause the efficiency anomaly was calculated it depended on angle of incidence in a smooth manner, suggesting the electric fields inside the chamber were fairly homogeneous and/or weak. A weak field would enable the same fraction of electrons from the sample to reach the reference counter from each polarisation state, given the angles of incidence were identical. Secondly, if cross coupling were to affect the modulation, it would do so least at small grazing incidence angles where the number of photoelectrons reaching the reference counter is less then 1% of the total; but this is precisely where we see the highest modulations.

The planar gold photocathode data shown in figures 3.5 to 3.9 are strongly consistent, both across absorption edges and between energy ranges showing a broad band polarisation sensitivity characterized by 13% modulation at 8° grazing incidence. The dependence on grazing angle is also as expected with high modulations at low grazing angles and zero modulation approaching normal incidence. The one thing that could cast doubt on these data is the possibility that the normal incidence position is poorly calibrated. As already stated, discrepancies of 0.65° and 1° in angular calibrations were found during the experiment. Angular miscalibration would lead to S and P measurements being conducted at slightly different angles of incidence, with the likelihood we would be measuring differences in quantum efficiency caused by differing angles of incidence rather than by any polarisation dependence. Worse still, angular miscalibration would also have an angular dependence similar to figure 3.5 as a small angular offset would become increasingly dominant at low grazing angles. Figure 3.3 does however show that any such error in the S measurement angle is only ~ 0.1°, far below the level needed to explain the modulations seen above, although the error in the α angle for P was less accurately defined. Thus, although the data are very promising, one would only be able to definitely state whether a polarisation dependency in X-ray photoemission exists by conducting S and P measurements with all angles...
defined to within 0.1° or fixed as in the angled plate measurement.

We have also shown that searches for polarisation sensitivity on CaI in the normal synchrotron flux regime are futile and if the X-ray vectorial effect were demonstrated, experiments on this higher quantum efficiency material should be conducted using a lower flux, polarised source.
Chapter 4

Polarimetric Measurements in the Energy Range 50 to 1000 eV

4.1 Experimental Details

A radically different experimental technique was adopted for this series of synchrotron measurements, concentrating on producing "unchallengable" results for a single grazing angle and single material - gold. Two important priorities were defined; (i) measurement of full modulation curves (0° - 360° in azimuth) at fixed grazing incidence angle, (ii) to attempt to understand the physics of the process by using an electron energy analyser to examine any variation in yields of primary and secondary electrons with azimuthal angle. Subsidiary investigations such as the variation of modulation with angle of incidence and measurements on different photocathode materials were discontinued, to minimise the likelihood of complications.

To measure a modulation curve at a number of azimuthal angles, ϕ, requires the photocathode to be rotated about the beam, preferably through a full 360°. The modulation signature should then follow a sin 2ϕ curve; the modulation factor, M, can be calculated from the strength of the 2ϕ component in the fourier analysed curve. Any misalignment between the beam axis and the axis of rotation would superimpose a sin ϕ component, a component which would be removable by fourier analysis and thus not compromise the data. Instead of trying to produce a rotating
experiment inside a fixed vacuum chamber we elected to use a rotating chamber, which had been originally designed to measure the polarisation of SRS beamline 5U.1. The rotating chamber allows electron spectroscopy to be conducted at a constant angle to the photocathode for all $\phi$ angles. It also has the advantages that internal cabling is not required to rotate through 360°, where there is always the risk it could obstruct the beam. Finally, construction of a stable experimental platform, which does not suffer deflection under rotation, is easier.

As the rotating chamber had been designed to fit beamline 5U.1, this line was an obvious choice for the experiment. This choice of beamline also had the important benefit of bringing variable polarisation to the study (section 4.2). Beamline 5 uses an undulator magnet, rather than the usual bending magnet as the photon source, and it is this, as described in section 1.3.4, which creates the harmonic structure in the polarisation and flux responses as shown in figure 1.10. The optical elements of beamline 5U.1 are shown in figure 1.11 and described in section 1.3.4. The beam is focused to a spot 1mm by 1mm, one metre from the focusing mirror. The rotating chamber had been designed so as its centre laid at the focus in the absence of a beam monitoring section, which thus also had to be removed for our experiment. A new beam monitor was therefore located inside the experimental chamber as illustrated in figure 4.1.

The chamber was rotated by means of a hand driven worm wheel arrangement, with full
360° rotation possible. In principle continuous rotation is possible, but in practice this was not used due to the number of cables attached to the chamber. The photocathode consisted of a 780Å layer of gold vacuum evaporated at Leicester onto a polished stainless steel substrate. The photocathode was then mounted alongside a quadrant anode (below) on a linear drive, spacers being used to ensure the centres of both the photocathode and quadrant anode were at the same height. The beam monitor used was a piece of high transmission copper micromesh, 250 cells per inch, chosen to have no absorption edges in the required energy range and a cell size so small that beam position with respect to the mesh is unimportant.

Two quadrant anodes manufactured in Leicester were included in the chamber to permit the correct axial alignment of the chamber. A quadrant anode consists of four gold coated stainless steel square sections separated by 100μm in a window-pane arrangement. The sections are electrically insulated from one another but are connected, through a fixing screw and four way electrical feedthrough, to current amplifiers. When the X-ray beam is centred on the anode the signal from all four quadrants will be identical. One quadrant anode shares a linear drive with the beam monitor at the front of the chamber, while the other is mounted alongside the photocathode at the chambers centre. Note that at the first quadrant anode the beam is not circularly symmetric (in fact ~ 3mm high by 11mm wide) which will lead to some error in centring.

A biased metal plate was also included above the photocathode to ensure that electrons from the photocathode could not reach the beam monitor as in previous experiments (see Chapter 3). To remove any contamination from the photocathode surface an argon ion gun was mounted on the vacuum chamber. A relatively low voltage (600 - 1000V) and short cleaning time (10 minutes) were used to avoid too much argon being embedded in the photocathode. Normally samples are heated after argon cleaning to drive off any argon, but no heater was available for this experiment. A cylindrical mirror analyser (CMA) was to be installed to look at the variation of primary and secondary photoyields as a function of azimuthal angle. However, this analyser could only be mounted on one vacuum port, where it would block the operation of the argon ion gun. It was thus decided to leave the CMA off the chamber for the first period of the experiment and insert it later.

Chamber alignment was a two stage process. First, the axis of rotation had to be discovered
so that the photocathode and quadrant anodes could be placed on it. Second, the chamber had to be aligned so its axis of rotation was coincident with the X-ray beam. An inaccuracy in finding the axis of rotation of the chamber would lead to the beam spot precessing about the centre of the photocathode, with the danger that some of the flux will miss the photocathode at certain azimuthal angles, creating a false modulation signal. The true axis of rotation was found with the chamber off the beamline to enable an optical alignment telescope to be mounted approximately on the axis of rotation and pointed at both quadrant anodes. The quadrant anodes were initially positioned approximately at the chamber centre by measurement of distance from the chamber walls. The telescope was then focused on the centre of the first anode and the chamber rotated to see how the anode centre moved. The anode was then moved appropriately and the procedure repeated until the anode centre moved less than 0.1mm under chamber rotation. The procedure was then repeated for the second anode.

Accurate alignment of the axis of rotation of the chamber to the beam axis is essential to reduce 360° modulation. With both quadrant anodes centred on the rotation axis the chamber was mounted on the beamline and pumped down to ~ 3.10^-8 mbar. The chamber was then rotated so that the linear drives would move the quadrant anodes in a horizontal plane and the position of the chamber adjusted so as to centre the beam. The chamber was held in a frame laterally adjustable in all three planes; vertically, horizontally along the beam direction and horizontally at right angles to the beam. The chamber was then rotated by 90° and the procedure repeated to find the vertical position. Repeating this procedure twice more, noting that adjustments nominally in one plane may also affect the other, ensured the beam was aligned to the chamber rotation axis.

### 4.2 Measurement Technique

The measurement technique relied partly on the unique properties of the undulator magnet source. The flux and degree of linear polarisation of the undulator output depend on both the monochromator energy setting and spacing of the undulator magnets (which vary the magnetic field \( B_o \) and thus \( K \) in equation 1.5). Figure 1.10a shows output flux as a function of energy and figure 1.10b shows the beam polarisation versus energy, both taken for a fixed undulator

69
The first harmonic can be tuned between 60 and 150 eV by variation of the undulator spacing. At the first harmonic, figure 1.10b shows an almost fully polarised beam is achieved, while tuning off the harmonic produces a 40-50% polarised beam. By tuning the undulator we were thus able to make modulation scans with different degrees of beam modulation, while all other factors remain identical including the X-ray energy. This provided a powerful check that any modulation recorded was due to a real polarisation dependence of the photocurrent rather than a spurious instrument artifact.

A typical measurement in the photon energy range 65 to 160 eV thus consisted of selection of the required monochromator energy and then adjusting the undulator spacing to position the peak of the first harmonic at the required energy. This was achieved by getting the Daresbury control centre staff to adjust the undulator spacing whilst we used the reference counters to monitor the flux, seeking the undulator spacing that gave maximum flux down the beamline. The replacement current to the photocathode was then measured for 15° azimuthal intervals to build up a full 360° modulation scan. The undulator spacing was then altered to find the flux and polarisation trough between the first and second harmonic, the energy, defined only by monochromator setting, being the same and a second modulation scan executed.

The range of undulator spacings only permits this technique to be used for photon energies below 160eV; above this limit the first harmonic is unobtainable. The degree of spurious modulation should, however, be quantified by the lower energy measurements so that modulation scans in just the high polarisation state could be conducted at a range of energies up to 1keV.

### 4.3 Total Electron Yields

Measurements were conducted for high and low beam polarisation states at 65eV, 70eV and then in 10eV steps up to 160eV. A range of high polarisation state measurements were conducted between 180 and 1000 eV, with measurements a few eV either side of the gold N edges at 335, 353, 546, 643 and 762eV.

Figures 4.2 and 4.3 illustrate the variation of photocathode quantum efficiency with azimuth angle - so called "modulation scans". Absolute efficiency calibration was not attempted due
Figure 4.2: Full modulation curves at 65eV and 8° grazing incidence. The photon beam is P polarised at azimuthal angle, $\phi=0^\circ$ and S polarised at $90^\circ$. $\times$: data in $15^\circ$ azimuthal steps. Full curve: Best fit $\sin(2\phi + \omega)$ curve. Top) ’100%’ linearly polarised beam producing a modulation factor of $0.078 \pm 0.002$ and bottom) ’50%’ linearly polarised beam producing a modulation factor of $0.032 \pm 0.002$. 

71
Figure 4.3: Modulation curve and best fit $\sin (2\phi + \omega)$ representation for the fully polarised beam (above) where $M = 0.035 \pm 0.004$ and for a 50% polarised beam (below) where $M = 0.010 \pm 0.003$. Photon energy 110eV, angle of incidence $8^\circ$. 

72
to second and higher order beam contamination, which accounted for up to 25% of the flux at 60eV, coupled with carbon and oxygen contamination of the beam monitor. The beamline was however very stable and good relative measurements were all that were required. The relative efficiency values presented are calculated by dividing the drain current from the sample by the drain current from the reference counter and then corrected so as to average unity across the range $\phi = 0^\circ \cdot 360^\circ$. The reference counter readings were uncorrected for its efficiency at the photon energy and its transmission properties were disregarded.

Measurements for both high and low polarisation states are displayed in figures 4.2 and 4.3, taken at 65eV and 110eV respectively. A significant $\sin 2\phi$ structure characteristic of polarisation dependence can be seen in all figures, with an instrumental $\sin \phi$ structure becoming increasingly prominent in the low polarisation state plots. As can be seen the $\sin \phi$ component would produce a significant inaccuracy in a modulation calculated by equation 2.1. Modulation factors were calculated for this experiment by three more accurate methods. Firstly from the strength of the $\sin 2\phi$ component in the Fourier transform, from a best fit $\sin 2\phi$ curve fitted to the data and by a linear least squares fit to a plot of $\cos(2\phi)$ versus relative efficiency. The three values were then averaged, and it is these modulation factors, for different beam polarisation states and energies, that are tabulated in table 4.1 and quoted elsewhere in this chapter. Note that modulation factors calculated in this manner are always positive; P state dominance, designated previously by a negative modulation factor, here produces an offset phase angle of approximately zero. S state dominance produces phase angles of $\pm 90^\circ$.

In general terms, the recorded modulation factor decreases with energy above 65eV, with no significant modulation above ~150eV. Measurements taken between 180eV and 1000eV also exhibit very low modulation values, with the gold N edges in the range having no effect. Note that Kunz (1979b) also found the normal incidence photoyield for an evaporated gold photocathode to be featureless in the 50 - 170 eV band. Interestingly the modulation reaches a minimum around 160eV before moderately increasing again, although with the polarisation state switching from P to S dominance. Figure 4.4 shows the azimuth scan at a photon energy of 1000eV, where the phase shift is noticeable when compared to that of figures 4.2 or 4.3.

The switch in polarisation state is unlikely to be of physical origin, with the "X-ray vectorial effect" and differences in the Fresnel reflectivity coefficients (below) both suggesting P dominance.
<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>High polarisation state modulation factor (%)</th>
<th>Low polarisation state modulation factor (%)</th>
<th>Phase Angle (high state)</th>
<th>Phase Angle (low state)</th>
</tr>
</thead>
<tbody>
<tr>
<td>65</td>
<td>7.8</td>
<td>3.2</td>
<td>1°</td>
<td>1°</td>
</tr>
<tr>
<td>70</td>
<td>7.2</td>
<td>3.0</td>
<td>0°</td>
<td>4°</td>
</tr>
<tr>
<td>80</td>
<td>6.8</td>
<td>3.0</td>
<td>3°</td>
<td>4°</td>
</tr>
<tr>
<td>90</td>
<td>5.6</td>
<td>2.8</td>
<td>-12°</td>
<td>-12°</td>
</tr>
<tr>
<td>100</td>
<td>4.7</td>
<td>1.6</td>
<td>-12°</td>
<td>-13°</td>
</tr>
<tr>
<td>110</td>
<td>3.5</td>
<td>1.0</td>
<td>-12°</td>
<td>-11°</td>
</tr>
<tr>
<td>120</td>
<td>2.6</td>
<td>-</td>
<td>-11°</td>
<td>-</td>
</tr>
<tr>
<td>130</td>
<td>1.9</td>
<td>0.3</td>
<td>2°</td>
<td>-15°</td>
</tr>
<tr>
<td>140</td>
<td>1.3</td>
<td>0.2</td>
<td>1°</td>
<td>47°</td>
</tr>
<tr>
<td>150</td>
<td>0.7</td>
<td>0.5</td>
<td>-3°</td>
<td>68°</td>
</tr>
<tr>
<td>160</td>
<td>0.4</td>
<td>0.7</td>
<td>-13°</td>
<td>-85°</td>
</tr>
<tr>
<td>240</td>
<td>0.9</td>
<td>-</td>
<td>-85°</td>
<td>-</td>
</tr>
<tr>
<td>440</td>
<td>1.2</td>
<td>-</td>
<td>83°</td>
<td>-</td>
</tr>
<tr>
<td>640</td>
<td>1.2</td>
<td>-</td>
<td>80°</td>
<td>-</td>
</tr>
<tr>
<td>850</td>
<td>1.5</td>
<td>-</td>
<td>77°</td>
<td>-</td>
</tr>
<tr>
<td>1000</td>
<td>2.0</td>
<td>-</td>
<td>76°</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 4.1: Modulation factors between 65 and 1000eV where measurements below 160eV were conducted on the first harmonic (nominally 100% polarised beam) and tuned off the harmonic (50±10% polarised beam), whilst at higher energies measurements were only conducted with high beam polarisation, although not on the first harmonic. The phase angle is defined as the angle between the first minimum in the modulation curve and a 0° azimuthal angle defined with respect to the beam axis.
Figure 4.4: Quantum efficiency versus azimuthal angle of a gold photocathode at 8° grazing incidence to a 1000eV linearly polarised photon beam. *s data points; full curve: best fit sine $2\phi$ wave.

Also the shape of the curve, in figure 4.4, has a complex $\phi$ dependence, more likely to be caused by instrumental effects than polarisation dependence. Comparing modulation factors from high and low beam polarisation states also suggests the existence of an instrumental modulation with S state dominance. Between 65eV and 110eV the modulation factor measured in the low beam polarisation state was only 41% that measured for high beam polarisation. Figure 1.10; graphs showing beam polarisation against energy and flux versus energy for beamline 5U.1, shows why this ratio is unlikely. The maximum and minimum beam polarisations available are 98% and 41% respectively, but the problem lies in finding the first harmonic and the polarisation trough between first and second harmonic. The only guide available as to the beam polarisation is the variation of the flux down the beamline with undulator spacing. The polarisation peak associated with the first harmonic is accurate to find in this way as the flux spike is very narrow, but the maximum in the beam polarisation is comparatively broad. The minimum in the flux spectrum is very broad, encompassing a region of over 10eV, which makes accurate positioning on the narrow polarisation minimum impossible. In fact, over the 10eV region of the flux minimum, beam polarisation varies from 41% to 65% so the beam polarisation for the above measurements were taken as $50 \pm 10\%$. Note that this assumes the beam polarisation minimum and the shape
of the polarisation response are independent of the undulator spacing and thus energy of the first harmonic. The absence of measurements or calculations makes this assumption unprovable.

Both the switch in polarisation states, the odd "shape" of the 240 to 1000 eV data and the apparent anomaly in the beam polarisation appear best explained by an instrumentally induced $S$ dominant $\sin 2\phi$ polarisation signature of strength 1 to 2% coupled with a $\sin \phi$ and higher components. If this is the case we can 'correct' the modulation scans by dividing them by the believed instrumental offset. Two possible methods exists for estimating the instrumental response, (a) taking a high energy data set where no true polarisation dependence is believed to exist, or (b) by normalising the relative efficiencies to average unity and applying:

$$\text{Instrumental Offset}(\phi) = 1 + Q_f(\phi) - 2Q_h(\phi)$$ (4.1)

where $Q_f(\phi)$ is the normalised efficiency for a full beam polarisation measurement at azimuthal angle $\phi$ and $Q_h(\phi)$ the equivalent for the 50% polarised beam. Both instrumental calibration methods lead to $P$ dominant modulations rising by approximately 1%, and some reduction in the $\sin \phi$ component. Figure 4.5 shows the result of comparing high and low beam polarisations to produce an instrumental calibration file, then used to correct the 65eV off-first-harmonic data originally displayed in figure 4.2. Note these data was not used in production of the calibration file. All experimental data and modulations quoted below have been corrected for instrumental response in this way.

The photocurrent from an X-ray photocathode is proportional to one minus the reflectivity, and the reflectivities for $S$ and $P$ polarised photons differ in this energy regime. This will thus produce a modulation given by (Fraser et al. 1991b)

$$M = \frac{(1 - R_s) - (1 - R_p)}{(1 - R_s) + (1 - R_p)} \approx \frac{R_p - R_s}{2 - R_s - R_p}$$ (4.2)

where $R_s$ and $R_p$ are the Fresnel reflectivities for $S$ and $P$ polarised X-rays as given by equations 2.9 and 2.10 respectively. Two methods of finding the complex dielectric constants on which these calculations depend were used. A theoretical approach was based on use of the Cromer and Liberman database (Cromer & Liberman 1970) of atomic and anomalous scattering factors from which the real and imaginary parts of the dielectric constant can be calculated. Measurements of the reflectivity and transmission properties of gold over the energy range 40eV - 1.3keV,
Figure 4.5: Quantum efficiency versus azimuthal angle of a gold photocathode at 8° grazing incidence to a 65eV linearly polarised photon beam; corrected for instrumental response. *s: data points, curve: best fit sin 2φ wave. The modulation factor of 0.044 compares to the uncorrected modulation factor of 0.032 (compare with figure 4.2 lower panel).

Conducted by Wolf et al. (1994), were also used to calculate the dielectric constant. The values obtained from transmission and reflectivity measurements were significantly different, so, on the advice of the author, the values obtained by transmission measurement were taken as the more accurate.

Figure 4.6 compares the modulation factor calculation from equation 4.2 with our experimental data over the range 60 to 180 eV. The measured modulations are somewhat lower than the theoretical predictions, an effect unlikely to be caused by instrumental misalignment. The experimental data, is though self consistent and of similar shape to the Fresnel modulation predicted from the data of Wolf et al. (1994). The Cromer & Liberman (1970) predictions show good numerical agreement above 100eV, but diverge strongly from measurements below this energy.

Various mechanisms to explain the apparent low energy divergence were considered. Surface roughness of the sample would be expected to have greatest effect for short wavelength radiation and can thus be discounted. The effect of any residual argon from ion cleaning was investigated by calculating the reflectivities for an argon-gold composite by the atomic scatter-
Figure 4.6: Comparison of experimental modulation data and predictions of the modulation due to differences in the Fresnel reflectivities of S and P polarised X-rays.

In common with any modulation measurement or calculation only small errors in reflectivities or efficiencies are required to produce significant inaccuracies in the modulation. As seen previously the two predicted modulations can hardly be said to strongly agree and a calculation with the reflectivity data of Wolf et al. (1994) would produce a third distinct prediction.

Although there exist slight differences in the experimental data and theoretical predictions the result strongly suggest that photocurrent polarisation sensitivity below 180eV is real and is
Figure 4.7: The effect of argon contamination of the photocathode surface. *s experimental modulation data. The full curve denotes the expected modulation from a pure gold surface; the dotted line from a surface contaminated with 5% argon and the dashed line with 20% argon contamination.

Figure 4.8: The effect of a constant error in the grazing angle of incidence of the photocathode to the beam. *s experimental data taken at nominal 8° grazing incidence. The full curve illustrates the expected Fresnel modulation at 8°, while the dotted and dashed lines represent 6° and 10° respectively.
the result of differences in the S and P reflectivities. The data, however, is very different to that measured in our previous experiments in this energy range, which, as shown in figure 2.13, show apparent modulation factors for a gold photocathode of up to 40% at 150eV. The present photocurrent data must be regarded as more reliable due to the full azimuth scan and 30% polarised beam check, which calls into question the earlier results. No analysis of the cause of inaccuracies in the earlier measurements will be presented here, although the presence of sine $\phi$ instrumental modulation and an incorrectly calibrated normal incidence position are both possible. Questions must also be asked of the data presented in chapter 3 which was collected in a manner subject to similar errors, although it would be preferable to conduct more accurate measurements before concluding another polarisation dependent mechanism cannot be present at these higher energies.

4.4 Electron Energy Resolution

We had initially hoped to measure the spectral characteristics of the emitted electrons to aid our understanding of the physical mechanisms behind any polarisation dependency. This, however, proved impossible as the cylindrical mirror analyser we were to use was found to be heavily organically contaminated forbidding its use in the ultra high vacuum environment. This loss, given the general agreement of the data with a reflectivity based mechanism, was not catastrophic.
Chapter 5

X-ray Vectorial Effect Modelling

High modulations have been seen in the primary photoyields of silicon at 2.69keV, where Shaw et al. (1994) measured modulation up to 40% when all secondaries and some low energy primaries were rejected by a biasing potential. A model of the physical mechanisms underlying this experiment was developed to see if the primary photoemission mechanism could yield a polarimeter with advantages over those currently available. This model was based on an existing model of electron transport within a Charge Coupled Device (CCD) pixel polarimeter (Holland et al. 1995). Such a polarimeter utilises differences in the spread of multi-pixel events in orthogonal directions when the electric vector of a normal incident X-ray is parallel to one of these directions.

5.1 Polarisation Sensitive Primary Photoemission Model

The Monte Carlo model is based on primary photoemission from a silicon photocathode, silicon being the material used for the Shaw et al. (1994) experiment. A range of energies and grazing angles was chosen to include 2.69keV and 5°, (the values used by Shaw et al. (1994)) and normal incidence, where, by definition, the polarisation sensitivity must be zero. An X-ray beam, generated and then polarised by Bragg reflection is simulated by producing a normal distribution in energy centred on the chosen energy, $E_\gamma$, and with a fwhm of approximately 0.001$E_\gamma$. Test energies were limited to the band above the silicon K edge (1839eV), where absorption occurs
Figure 5.1: The coordinate systems used by the polarisation sensitivity model. The (x,y,z) axes are defined with respect to the sample surface (which lies in the (x,y) plane) and the (x',y',z') coordinates are defined with respect to the X-ray path.

The initial energy of the photoelectron is then the X-ray energy minus the binding energy of the shell the X-ray was absorbed in.

A coordinate system (x',y',z') based on the incident X-ray is defined such that z' is antiparallel to the X-ray path and the photon's electric vector \( \varepsilon \) lies in the (x',y') plane as shown in figure 5.1. A further set of orthogonal planes (x,y,z) are defined such that z is vertical and the sample surface is in the (x,y) plane. The angle of incidence, \( \theta \), then corresponds to the angle between z' and the (x,y) plane. The model simulates the absorption of an X-ray at a depth chosen randomly from the exponential distribution given by equation 5.1

\[
z = -\frac{\sin \theta \log(r_1)}{\mu(E_x)}
\]  

(5.1)

where \( \mu(E_x) \) is the linear absorption coefficient in silicon for an X-ray of energy \( E_x \), \( \theta \) is the grazing angle of incidence and \( r_1 \) a random number between zero and unity. We are only interested in whether the produced photoelectron escapes the surface or not so no (x,y) positions are stored.

The direction of the photoelectron is then chosen at random from an angular distribution.
\[ \sin^2 \psi \cos^2 \varphi \text{ where } \psi \text{ is the angle between the incident photon direction and the photoelectron ejection direction and } \varphi \text{ is the angle between the electron and the electric vector, } \varepsilon. \] This distribution is appropriate for X-ray absorption by electrons in the spherically symmetric s shells, but not for absorption by shells of non zero angular momentum which, as shown by Afans'ev et al. (1988), exhibit less polarisation sensitivity. For X-ray energies above the Si K edge, absorption in asymmetric shells occurs in approximately 1% of cases and for simplicity was thus ignored.

In practice, the angular distribution is generated by randomising the electron's direction vector in terms of \( \alpha' \) and \( \beta' \) where \( \alpha' \) is the angle between the photoelectron and the \((x',y')\) plane and \( \beta' \) the angle between the projection of the photoelectron onto the \((x',y')\) plane and \( x' \). These are then found by generating two random numbers \((r_2 \text{ and } r_3) \) in the range \([0,2\pi]\), then stepping through \( \alpha' \) and \( \beta' \) values until the following conditions are independently satisfied:

\[
\begin{align*}
\alpha' &= r_2 < -\sin(\alpha') \cos(\alpha') \quad (5.2) \\
\text{and} \\
\beta' &= r_3 < -\sin(\beta') \cos(\beta') \quad (5.3)
\end{align*}
\]

This assumes that the electric vector \( \varepsilon \) lies along the \( x' \) direction, which is the case for the P polarisation state. For the S polarisation state, the \( \beta' \) values obtained from equation 5.3 have 90° added to them to simulate \( \varepsilon \) actually lying along the \( y' \) direction. The photoelectron direction vector is then transformed into \((x,y,z)\) co-ordinates by the matrix transformation:

\[
\begin{pmatrix}
\hat{x}' \\
\hat{y}' \\
\hat{z}'
\end{pmatrix} =
\begin{pmatrix}
\cos(90 - \theta) & 0 & -\sin(90 - \theta) \\
0 & 1 & 0 \\
\sin(90 - \theta) & 0 & \cos(90 - \theta)
\end{pmatrix}
\begin{pmatrix}
\hat{x} \\
\hat{y} \\
\hat{z}
\end{pmatrix}
\]

\[(5.4)\]

where \((90 - \theta)\) is the angle of incidence measured from the normal to the cathode surface and

\[
\begin{pmatrix}
\hat{x}' \\
\hat{y}' \\
\hat{z}'
\end{pmatrix} =
\begin{pmatrix}
1 \\
\tan(\gamma') \\
\tan(\gamma') / \tan(\beta')
\end{pmatrix}
\]

\[(5.5)\]
The angle, $\alpha$, between the photoelectron direction and the $(x,y)$ plane, is then calculated from

$$\alpha = \tan^{-1} \left( \frac{\sin \theta \cos \beta + \cos \theta \tan \alpha'}{\left( \cos^2 \theta \cos^2 \beta' - \sin \theta \tan^2 \alpha' - \sin \theta \cos \theta \tan \alpha' \cos \beta' + \sin^2 \beta' \right)^{\frac{1}{2}}} \right)$$  \hspace{1cm} (5.6)$$

Physically, the distribution of $\alpha$ values should exhibit mirror symmetry about the X-ray trajectory. Equation 5.6 does not produce this result, yielding only values between 0° and 180° or ± 90° dependent on implementation. Adding 180° to a randomly selected half of the $\alpha$ values corrects this anomaly.

Apart from the photoelectrons, a number of Auger electrons are also created by X-ray absorption. For 96% of X-ray absorptions in the K shell an Auger electron is released; in the remaining 4% of cases relaxation occurs by K shell fluorescence; these photons are assumed not to be re-absorbed in the sample. Auger electrons are emitted with an isotropic spatial distribution; the angle $\alpha$ is then found from;

$$\alpha = \cos^{-1}(1 - 2r_4)$$  \hspace{1cm} (5.7)$$

Equation 5.7 is derived from consideration of the surface area of a sphere for which the polar angle lies between $\alpha$ and $\alpha + \delta \alpha$ and integrating to find the fractional area. A random number, $r_4$, between zero and unity is taken to represent the fractional area from which $\alpha$ can then be calculated. The probabilities of each possible Auger transition for K shell absorption in silicon are as given in table 5.1. On creation of a L shell vacancy, Auger emission creates a 132eV electron, but its probability is only 2.5%; the vast majority (97%) of the time a Coster-Kronig rearrangement occurs which releases an electron with an energy of only 49.5 eV (Fraser et al. 1994). Neither of these effects will thus lead to the emission of high energy electrons, and as such are ignored.

Both the primary photoelectron and Auger electrons will free secondary electrons as they pass through the material. In this model the secondary electrons are not tracked, but the loss of energy from the primary and Auger electrons is fully taken account of. The experiments described in chapter 4 and by Shaw et al. (1991) demonstrated that little or no polarisation information was retained by the secondary electrons. In any design of a primary photoelectron polarimeter the secondaries that escape the sample must be discriminated out by a repelling
<table>
<thead>
<tr>
<th>Auger Transition</th>
<th>Energy (eV)</th>
<th>Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>KL₁L₁</td>
<td>1542</td>
<td>0.192</td>
</tr>
<tr>
<td>KL₁L₂₃</td>
<td>1591</td>
<td>0.389</td>
</tr>
<tr>
<td>KL₂₂L₂</td>
<td>1641</td>
<td>0.273</td>
</tr>
<tr>
<td>KL₁M₁</td>
<td>1679</td>
<td>0.072</td>
</tr>
<tr>
<td>KL₁M₂₂</td>
<td>1685</td>
<td>0.0032</td>
</tr>
<tr>
<td>KL₂₂M₁</td>
<td>1728</td>
<td>0.100</td>
</tr>
<tr>
<td>KL₂₂M₂₂</td>
<td>1735</td>
<td>0.0042</td>
</tr>
<tr>
<td>KM₁M₁</td>
<td>1816</td>
<td>0.0066</td>
</tr>
<tr>
<td>KM₂₂M₂₂</td>
<td>1822</td>
<td>0.00099</td>
</tr>
</tbody>
</table>

Table 5.1: Energies and Probabilities for the various Auger transitions in Silicon (Fraser et al. 1994)

Each primary or Auger electron is then followed as it undergoes elastic and inelastic collisions until its energy is less than 25eV or it escapes the surface. For simplicity, the individual inelastic processes (valence and core-level ionisation, plasmon and phonon interactions) were represented by a collective mean free path, $\lambda_{\text{inel}}$ and assigned a characteristic energy loss per collision derived from the inverse mean free path and the stopping power curves of Tung, Ashley & Ritchie (1979). The treatment of elastic scattering of electrons is based on that of Cengiz & Ozmutlu (1994) corrected as in Holland et al. (1995). The total elastic scattering mean free path $\lambda_{\text{el}}(E)$ is derived from the screened Rutherford formula with an unscaled screening factor $\beta_N$. According to the data of Adesida, Shimizu & Everhart (1980) the unscaled screening factor produces the best approximation for electron energies of order 1keV. Above 5keV a scaling factor of 0.48 is more applicable, but for reasons of continuity was not implemented. The effect of this will be minor, as the underestimation of the number of elastic scattering events is offset by an overestimation of the scattering angle, with a larger $\beta_N$ in equation 5.10. Table 5.2 collates the mean free paths and characteristic energy loss for electrons of various energies in silicon.

The combined mean free path given by:

$$\frac{1}{\lambda} = \frac{1}{\lambda_{\text{inel}}} + \frac{1}{\lambda_{\text{el}}}$$  \hspace{1cm} (5.8)
Table 5.2: Values of elastic and inelastic scattering mean free paths and inelastic scattering energy loss as a function of electron energy, $E$

<table>
<thead>
<tr>
<th>Electron Energy ($E$ eV)</th>
<th>Energy Loss ($\Delta E$ eV)</th>
<th>$\lambda_{\text{el}}$ ($\text{Å}$)</th>
<th>$\lambda_{\text{in}}$ ($\text{Å}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>23</td>
<td>4</td>
<td>5.4</td>
</tr>
<tr>
<td>200</td>
<td>30</td>
<td>7</td>
<td>9.5</td>
</tr>
<tr>
<td>500</td>
<td>29</td>
<td>11</td>
<td>21.8</td>
</tr>
<tr>
<td>1000</td>
<td>41</td>
<td>20</td>
<td>42</td>
</tr>
<tr>
<td>2000</td>
<td>45</td>
<td>33</td>
<td>83</td>
</tr>
<tr>
<td>5000</td>
<td>49</td>
<td>70</td>
<td>206</td>
</tr>
<tr>
<td>10000</td>
<td>50</td>
<td>125</td>
<td>509</td>
</tr>
</tbody>
</table>

while the actual path length travelled between collisions, $S$, is found from

$$S = -\ln(r_5)$$  \hspace{1cm} (5.9)

where $r_5$ is a uniform random number between 0 and 1. Another independently generated random number, $r_6$, is used to determine whether that collision is elastic or inelastic. If $r_6 < \frac{1}{\lambda_{\text{el}}}$ then it is elastic, otherwise the collision is taken to be inelastic.

In the case of an elastic collision the azimuthal scattering angle, $\gamma$, is $2\pi r_7$ and the polar scattering angle, $\alpha$, is given by;

$$\cos \alpha = \frac{1 + \beta_N - r_8(1 + 2\beta_N)}{1 + \beta_N - r_8}$$  \hspace{1cm} (5.10)

where $r_7$ and $r_8$ are further uniform random numbers between 0 and 1 and $\beta_N$ the atomic screening factor (not to be confused with the angle $\beta'$). This is given by Nigam, Sundaresan & Wu (1959), as;

$$\beta_N = 5.43 \frac{Z^2}{E(\text{eV})}$$  \hspace{1cm} (5.11)

For silicon with atomic number $Z=14$, equation 5.11 becomes $\beta_N = 31.54/E$. In the case of an inelastic collision, the azimuthal scattering angle remains $\gamma = 2\pi r_7$, but the polar scattering angle, as given by Shimuzu & Ichimura (1983), is found from
After either type of interaction the electron depth and new value of $\alpha$, the angle between the $(x,y)$ plane and the electron are updated by

$$\alpha_{i+1} = \alpha_i + \gamma \sin \gamma$$

and

$$z_{i+1} = z_i - S \sin \alpha_i$$

In the case of an inelastic collision the electron energy is also decremented by $\Delta E$. The process is then repeated until either the electron dissipates virtually all its energy or escapes the surface. Those electrons which are ejected from the sample have their emission energies recorded to obtain energy spectra for comparison with experiment.

5.2 Results from the Primary Photoemission Model

Figure 5.2 shows the calculated quantum efficiency versus discriminator setting for 2.69 keV X-rays incident to silicon at a 5° grazing angle. This models the physical effect of a varying the potential between the silicon photocathode and the electron detector. Only electrons with energies greater than the discriminator setting are hence included in the quantum efficiency calculation. Any photocathode polarimeter would operate in this manner with a discriminator voltage optimally set for the range of X-ray energies of interest.

The quantum efficiency (with 25V discriminator voltage to block the detection of secondaries) of such a detector can be seen to be 3.3 % at 2.69 keV. Three distinct regions can be made out in the graph. Above the highest Auger energy (1822eV discriminator setting) the efficiency is due solely to L shell photoemission. Between the Silicon K edge (~ 850V repelling voltage) and 1822eV the efficiency is due to both Augers and L shell photoelectrons. In the low energy region, below 850 eV, emission comes from both K and L shell photoelectrons and Auger electrons. Note that for all discriminator voltages the P polarised yield dominates.

Figure 5.3 shows the measurements by Shaw et al. (1994) of quantum efficiency versus discriminator voltage of a silicon sample to S and P polarised 2.7keV photons. In general appearance,
the predictions of figure 5.2 and the measurements closely resemble one another, both exhibiting the same three-stage structure and differences between the efficiencies of S and P polarisation states particularly below ~ 850V. The possible effect of oxygen contamination of the silicon sample was recognised by Shaw et al. (1994) by the inclusion of the position of the oxygen edges. There does, however, exist a significant disagreement between the magnitude of this polarisation sensitivity between model and experiment. This is better illustrated by figure 5.4 which compares the modelled (a) and experimental (b) modulation factor versus repelling voltage. In both cases the modulation has been calculated by equation 2.1 from the efficiency data of figures 5.2 and 5.3. Note the experimental data extends to 3000eV, beyond the supposed 2.69 keV photon energy. This is likely the result of a significant second order component in the beam.

The modulation above 1822eV (i.e. L shell photoemission) is about 35 % experimentally and modelled to be only 20 - 30 %. The discrepancy between experiment and model for K shell photoemission is even greater, 8-10 % compared to a predicted values of only 4%. Assuming the experimental results are accurate, it would appear the model is imperfect at low electron energies. The disagreement is certainly lower at higher electron energies: the electron interaction part of
Figure 5.3: Experimental photoemission results from a Si photocathode as a function of repelling voltage for both S and P polarisation at 5° grazing incidence (Shaw et al. 1994).

Our model has been used to model CCD polarimetry at electron energies above 10keV where the fit to experiment is far better (Holland et al. 1995). A possible reason for this is inaccuracy in the scattering angles at low energies, with the elastic scattering angles being the least dependable. An upper limit on possible modulation could be modelled by setting the scattering angle for elastic collisions to be zero. The results of which as displayed by the upper curve (□’s) in figure 5.4 shows not even this limiting assumption can quite reproduce the experimentally observed modulation, although the disagreement is no longer energy dependent. Alternatively setting the inelastic scattering angles to zero also increased the modulation, but to a lesser extent, as the inelastic scattering angles are considerable smaller than the elastic scattering angles anyway.

The primary photoelectric efficiencies (electrons per photon) produced by the model can also be compared to experimental data. Measurements on a silicon (Z=14) photocathode are not available, but Eliseenko, Shchemelev & Rumsh (1968) conducted measurements on aluminium (Z=13) bombarded by 1.74 keV to 8keV X-rays with a 50V repelling grid to block the detection of secondary electrons. Figure 5.5 compares experimental and modelled results over this energy range at 20° grazing incidence.
Figure 5.4: a) Modelled results for 2.69keV X-rays at 5° grazing incidence with the model described in section 5.1 (+) and the same model's upper limit on modulation produced by assuming no elastic scattering (□); b) Experimental Modulation data of Shaw et al. (1994) also at 2.69keV and 5°.
Figure 5.5: Comparison of primary quantum efficiency predictions for a silicon photocathode at 20° (+'s) and at 5° scaled to 20° by the approximate cosec θ dependence (curve), with the aluminium quantum efficiency measured at 20° grazing incidence (o's).

Due to the length of time taken to run the model software (40 hours on a Sparc Station 10), only four calculations at 20° grazing incidence were made. To illustrate the likely shape of this curve efficiency calculations for 5° grazing incidence were divided by a factor of 3.924. The factor, a result of dividing \( \sin 20° \) by \( \sin 5° \) assumes the quantum efficiency of a photocathode is roughly dependent on the cosec of the grazing angle (Fraser 1983b). This relationship follows from the simple geometric consideration that at larger grazing angles more photons will be absorbed deeper in the sample; that the 5° and 20° points do not agree exactly shows that the cosec law is only an approximate relation. Agreement between the modelled and experimental results is reasonably good at high energies. In fact it would be hard to say whether the differences were due to errors in the model, experimental errors or as a result of differences between silicon and aluminium. At lower photon energies, however, we again see a marked disagreement, reinforcing the suggestion that the electron transport model below a few keV is not ideal.

At 2.69 keV the model and experiment show that differences in the primary photoelectron production efficiency between the two polarisation states would not be useful in producing an efficient, high modulation X-ray polarimeter. Fixing the discriminator voltage forces one
Figure 5.6: Predicted modulation factor and primary quantum efficiency versus discriminator voltage for 4keV photons at 5° grazing incidence. Modulation is represented by the triangles and the right hand scale, with quantum efficiency for S (O's) and P (o's) polarised X-rays on the left hand scale.

to choose either high modulation with a very low quantum efficiency or low modulation with moderate quantum efficiency. Our model can, however, allow us to see how such a detector would respond to other photon energies. At 4keV (figure 5.6) the primary photoeffect becomes more useful as a high modulation with an almost decent quantum efficiency (~ 1%) becomes available. This is a result of the possibility of removing the polarisation-insensitive Auger electrons by use of an appropriate discriminator voltage, while maintaining the collection of both K and L shell photoelectrons. A discriminator voltage of 1822eV blocks the detection of all Augers; the K and L shell photoelectrons above this energy have a modulation factor of approximately 20% with a quantum efficiency of 1%. Above approximately 2100eV, only L shell photoelectrons are present, and thus the quantum efficiency has dropped to only a few tenths of a percent and the modulation is also lower, but rises sharply with higher discriminator voltages. Higher energy photoelectrons, from the same shell, will have undergone fewer scattering events in the photocathode and thus retained more of their original polarisation information, leading to the energy dependent modulation seen in both K and L shell photoelectrons.
Figure 5.7: Predicted modulation (triangles) and quantum efficiencies of $S$ (□'s) and $P$ (○'s) polarised 7keV photons versus repelling potential. Grazing angle - 5°.

Figures 5.7 and 5.8 show the corresponding curves for 7 keV and 19keV photons, illustrating the broad band capability of the detector. To obtain a high modulation of order 20% the discriminator voltage must, however, be higher to only measure the least scattered K shell photoelectrons. Selecting the best discriminator setting is thus a compromise for any broad band X-ray source, including all astronomical sources.

Modulation and efficiency data obtained at a range of photon energies are summarized in Table 5.3. For each energy an optimal discriminator setting has been calculated and the polarimetric figure of merit (as defined by equation 2.4) found for those settings and compared to that of the Thomson polarimeter modelled by Landecker (1972). The silicon polarimeter can be seen not to be competitive with the Thomson polarimeter, remembering that the ideal polarimeter has a figure of merit of unity. Worse, this is not really a fair comparison as the data for the Thomson polarimeter is for a polarimeter optimised at 14keV whereas the primary photoeffect polarimeter has, at this point, been optimised for each photon energy separately. Figures 5.9 and 5.10 plot the variation of modulation factor and the figure of merit over a range of X-ray energies for several fixed discriminator voltages.
Figure 5.8: Predicted modulation (triangles) and quantum efficiencies of S (□'s) and P (○'s) polarised 19 keV photons versus repelling potential for 5° grazing incidence.

![Graph showing modulation and quantum efficiency vs discriminator voltage](image)

Table 5.3: Variation of modulation factor and primary quantum efficiency with photon energy and discriminator setting; the comparison of the figure of merit (eqn 2.1) for a silicon photocathode and a Thomson scattering polarimeter. M denotes the modulation factor, \( \chi \), the quantum efficiency and \( \Psi \) the figure of merit; The subscript \( _0 \) denotes zero repelling voltage and \( _B \) the optimal voltage for that photon energy. The best voltage, \( V_B \) being calculated to minimise the figure of merit.

<table>
<thead>
<tr>
<th>X-ray Energy (eV)</th>
<th>Zero Repelling Voltage</th>
<th>Optimal Repelling Voltage</th>
<th>Polarimeter Figure of Merit</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( M_0 )</td>
<td>( \chi_0 )</td>
<td>( M_B )</td>
</tr>
<tr>
<td>2690</td>
<td>0.036</td>
<td>0.0343</td>
<td>0.036</td>
</tr>
<tr>
<td>4000</td>
<td>0.063</td>
<td>0.0232</td>
<td>0.174</td>
</tr>
<tr>
<td>6000</td>
<td>0.079</td>
<td>0.0174</td>
<td>0.164</td>
</tr>
<tr>
<td>8000</td>
<td>0.078</td>
<td>0.0149</td>
<td>0.163</td>
</tr>
<tr>
<td>10000</td>
<td>0.074</td>
<td>0.0135</td>
<td>0.144</td>
</tr>
<tr>
<td>15000</td>
<td>0.057</td>
<td>0.0121</td>
<td>0.133</td>
</tr>
<tr>
<td>20000</td>
<td>0.044</td>
<td>0.0119</td>
<td>0.112</td>
</tr>
</tbody>
</table>
Figure 5.9: Variation of modulation factor with photon energy for various marked discriminator energies.

Figure 5.10: Polarimetric figure of merit (as defined by equation 2.4) versus X-ray energy for various marked discriminator energies.
The main reason for the poor figure of merit is the very low primary quantum efficiency of silicon. An improvement would be to use a detector consisting of many ultra thin foils arrayed behind one another when seen by the photon, but effectively side by side so the emitted electrons are free to reach the detector. A schematic of such a multi-foil polarimeter is shown in figure 5.11; with electrons emitted from both faces of each foil being detected, the quantum efficiency is effectively doubled. In practice, each foil would be more of a strip than square, with a width not much greater than the beam width, so as to allow the instrument length and the size of the electron detector to be as small as possible and to reduce detector noise, electrostatic and beam focusing problems.

Taking an extreme case of a detector consisting of one hundred foils, various foil thicknesses were tested, to minimise the figure of merit for detecting polarised 4keV photons. The efficiency and modulation-versus-discriminator characteristics of an optimal 120Å "foil" are presented in figure 5.12, where, a 1775 eV discriminator setting equates to a very respectable figure of merit of 8.4. Leaving aside the difficulty of manufacture and handling of foils of this thickness, other problems are apparent from the graphs of figure 5.12, namely that the modulation peak is narrow and centred on the unscattered K shell photoelectrons. This is to be expected as electrons from P polarised photons are likely to be created with a motion toward both surfaces, and a distance to the surface of order the escape length, whereas S state photoelectrons will more
Figure 5.12: Predicted modulation (triangles) and quantum efficiencies of $S$ and $P$ polarised 4keV photons versus repelling potential for a 120Å silicon foil.

probably undergo many scattering events before escaping. The probability of getting electrons with energy significantly below the no loss K shell photoelectron energy is therefore more likely for $S$ polarised photons than $P$, reducing the modulation for these discriminator settings or the bandwidth for a fixed discriminator voltage detector. In fact, the figure of merit for the optimised 4keV polarimeter described above would drop to 46 at 5keV and 352 by 6keV.

Deciding on the optimal foil thickness is thus a trade off between quantum efficiency and detector bandwidth. Two other foil thicknesses were tested, 400Å and 1000Å with somewhat better combined characteristics. Figures 5.13 and 5.14 illustrate the variation of the figure of merit with photon energy for various repelling voltages for an ensemble of a hundred 400Å and 1000Å foils respectively. In both case figures of merit comparable to the Thomson scattering polarimeter are achievable, but over significantly smaller bandwidths. The 400Å foils give figures of merit as low as 8.0 over the low photon energy range, whereas the 1000Å foils significantly improve detector bandwidth whilst still giving low figures of merit at higher photon energies.

The conclusion to this modelling effort is that a multi foil silicon detector would make a rea-
Figure 5.13: The variation of the polarimetric figure of merit with photon energy for several fixed discriminator voltages for a hundred 400Å thick foils.

Figure 5.14: The variation of the polarimetric figure of merit for a hundred 1000Å thick foils.
A reasonably good polarimeter, but not quite as good as the Thomson scattering polarimeter or as easy to manufacture. A one hundred foil polarimeter is just about practical; if the detector acceptance width were 1mm, enough to accommodate the focal beam for XMM mirror module, each foil would have to be 11.4 mm long for 5° grazing incidence. The largest MCP electron detectors (similar to the 10 cm long MCPs built for AXAF) could only detect electrons from 10 foils, which implies a hundred foil polarimeter would require an beam size of 0.1 mm, in turn requiring a high resolution telescope like AXAF. There are, however a number of factors which suggest that in practice a multi-foil polarimeter could be as useful a device as the Thomson polarimeter or possibly better. Firstly there is the possibility that future measurements could confirm that the model underestimates, by up to a factor of two, both modulation and quantum efficiency in the low energy regime. A multi-foil polarimeter could also be optimised to have a peak response in the 4 to 6 keV region, while the Thomson scattering polarimeter works best around 10 keV with very limited sensitivity below 6 keV. An increased sensitivity at lower energies is important as cosmic X-ray source fluxes are generally far higher around 4 keV, than between 10 and 20 keV. Using different materials to produce the foils would also alter the detector characteristics, but in the absence of measurements or models of electron transport it is difficult to say that any given material would improve the polarisation sensitivity. What would be expected though is that use of a lower Z material would shift the sensitive range to lower photon energies. Beryllium is a likely candidate, with a K shell energy of 0.1 keV (c.f. 1.839 keV for Si) and the additional advantage that there would be no Auger contribution in the emitted electron spectrum improving the detector modulation response. Another area of investigation should examine whether a solid state design would be practical to ease the manufacturing and operational problems without significant loss of sensitivity.

5.3 Summary: Polarimetry

The origin of the X-ray vectorial effect was attributed in chapter 2 to the polarisation dependence of the direction of emission of the the primary photoelectron, with at least some of this polarisation information being transferred to any secondary electrons created. The results presented in the previous chapter for the soft X-ray range, coupled with measurements by the Columbia Astrophysical Laboratory group at 2.7 keV, also described in Chapter 2, cast very serious doubt
on the existence of such a mechanism, and suggest that early measurements of polarisation sensi-
tivity at X-ray energies above a few hundred electron volts were solely the result of experimental inaccnracies. At sufficiently low X-ray energies, polarisation dependent reflectivities would have created a real polarisation sensitive response, but this was entirely swamped by experimental inaccuracies. The real modulations produced by Fresnel reflectivity and reported in Chapter 4 are too small to be of use for a practical X-ray polarimeter, although they are important in the UV and Extreme Ultraviolet (EUV) where MCPs are used as readouts for gratings which have polarised the incoming photon beam by reflection.

The main thing learnt from this work is the importance of accurate angular calibration, for instance a 1% error in modulation only needs a 0.2°miscalibration in either S or P measurements conducted at 10°grazing incidence. Equally importantly one needs to be certain the calibration technique is correct otherwise one can never be really certain polarisation sensitivity is genuine. Best practice must be to use two independent angular calibrations or better still design the experiment so a miscalibration will have other effects distinguishable from a polarisation response.

A good place to put this into practice and advance the cause of X-ray polarimetry would be in testing the predictions of the model presented above and extending it to other materials.
Chapter 6

Calibration of Microchannel Plate Detectors for the AXAF High Resolution Camera (HRC)

6.1 Introduction to MCPs

Microchannel Plates (MCPs) are essentially glass discs with a large number of tiny holes in them. MCPs have been manufactured with holes of either round or square cross section and pore diameters ranging from 6μm on an 8μm pitch up to 100μm diameters. For use as photon or particle detectors, both plate surfaces are coated by an nickel-based electrode material and a large (~ 1-2 kV) potential difference applied. Figure 6.1a follows the incident photon as it interacts with the top of the channel and generates secondary electrons which escape into the vacuum and are accelerated by the channel electric field. These electrons drift across the channel diameter and strike the opposite channel wall releasing more electrons, and the process is repeated. In this way, at the end of a long channel, a large cloud of electrons leave the plate to be collected by the readout element. Common MCP configurations involve a second or even third MCP, to further amplify the electron signal and to stop positively charged nuclei from moving up through the stack as shown in figure 6.1b. In this way, the output charge pulse typically consists of $10^6 - 10^8$ electrons.
Figure 6.1: a) Microchannel Plate Operation; b) Common MCP configurations; the chevron detector (left) and the Z stack detector (right).
MCPs were first developed as image intensifier elements for night vision devices. They are, however, sensitive to both charged particles and to a range of wavelengths from UV down to X-ray, and have found use in, for example, plasma diagnostics, fast oscilloscopes and accelerator mass spectrometers. The X-ray quantum detection efficiency of bare plates is low (~ 10%) (Fraser 1982). Increased efficiency is achieved by coating the top of the channel with an X-ray photocathode material, normally CaI or KBr. This technique has been shown to give a 5 to 10 fold enhancement in efficiency, dependent on angle and energy, over bare plates. For X-ray astronomy MCP lead oxide glass without any potassium or rubidium content has been developed to avoid noise induced by internal $\beta$ emission of $^{40}$K or $^{85}$Rb atoms (Fraser, Pearson & Lees 1987; Siegmund, Vallerga & Warglin 1988).

Such a high efficiency coupled with the improvements mentioned above and high spatial resolution imaging ($\Delta x \sim 25\mu m$) led to MCPs being used on a number of successful astronomical missions including the Einstein and EXOSAT X-ray observatories and the ROSAT Wide Field Camera (WFC) and High Resolution Imager (HRI). MCPs will also be used as the basis for the High Resolution Camera onboard the US AXAF observatory (Weisskopf et al. 1995). The remainder of this chapter describes the results of the first attempt at synchrotron calibration of the MCP detector for AXAF.

### 6.2 The Advanced X-ray Astrophysical Facility (AXAF)

The Advanced X-ray Astrophysical Facility is a large X-ray observatory due to be launched by NASA in 1998. Figure 6.2 shows the major components of the satellite. The High Resolution Mirror Assembly (HRMA) consists of four nested mirror pairs in the Wolter 1 configuration, consisting of a hyperboloid section behind a paraboloid mirror. The outer mirror has a diameter of 1.2m, and all four have a focal length of 10m. There are two focal plane instruments making up the Integrated Science Instrument Module (ISIM). The High Resolution Camera (HRC) as described by Kenter et al. (1993), consists of a pair of MCP detectors, is designed for high resolution imaging, grating readout and fast timing measurements, while the AXAF CCD imaging spectrometer (ACIS), described by Garmire et al. (1985) is capable of performing simultaneous imaging and medium resolution ($E/\Delta E \simeq 50$) spectroscopy. The Leicester X-ray astronomy
Figure 6.2: Schematic of the deployed AXAF (Weisskopf et al. 1995). The fully deployed AXAF is 13.8m long with a 19.5m solar array span and a 4500 kg mass.

group is co-investigator on the development of HRC.

Two deployable transmission grating spectrometers, located just behind the mirror assembly, are used on AXAF. The Low Energy Transmission Grating (LETG) is optimised for low energies (0.05 - 2 keV) and the High Energy Transmission Grating (HETG) for the energy range 1 to 10 keV. Spectral resolving powers \( \frac{\Delta E}{E} \) in the range 100 - 2000 can be achieved with good efficiency. The main readout element for LETG is a strip MCP detector (HRC-S) and a strip CCD detector (ACIS-S) for HETG although the imaging detectors can also be used as redundant grating readouts.

Detailed overviews of the capabilities of the various instruments and the scientific objectives of AXAF are available (Weisskopf et al. (1995) and references therein).
6.2.1 The High Resolution Camera as spectrometer readout (HRC-S)

The High Resolution Camera (HRC) consists of two MCP detectors; HRC-I is AXAF's main imaging instrument, a large area (10 x 10 cm) small pore size (10μm) CsI-coated microchannel plate detector. The HRC-S detector is a 300mm by 27mm three-section MCP also coated with CsI. It uses a semi solid state substrate charge detector. This hybrid read out grid, as described by Kenter et al. (1993), consists of a wire grid wound in the narrow (cross dispersion) direction and a photo-etched micro-strip grid in the long (dispersion) direction. This allows tilting of the two outside sections slightly toward the grating to better approximate to the Rowland circle.

A vital part of the AXAF pre-launch programme is the calibration of all detectors, mirrors and gratings. The remainder of this chapter concentrates on an initial synchrotron calibration of the energy response of the MCP detector HRC-S as readout for LETG. The soft X-ray quantum detection efficiency of a microchannel plate is a complicated function of angle of incidence (defined relative to the channel axis) and energy. Quantum efficiency measurements on MCPs have been made many times before (Fraser 1982; Fraser et al. 1984) but usually only at isolated X-ray line energies. This is acceptable for imaging MCPs as their intrinsic energy resolution is poor and thus fine detail in their energy response would cause insignificant inaccuracies in the broad band QE. In the case of a spectrometer readout, more detailed calibration is required to avoid absorption edge structure in the responses of the photocathode and underlying MCP lead glass being misinterpreted as astronomical emission and absorption features. The planar photocathode work presented in Chapter 7 has confirmed that regions near absorption edges exhibit a complex quantum efficiency versus energy behaviour with the presence of not just simple steps but white line features and periodic EXAFS.

The level of detail achievable from 1eV monochromator resolution demonstrated in the photocathode efficiency measurements make the synchrotron the ideal method of calibration across the whole of the intended HRC-S bandwidth. Pioneering MCP photocathode measurements at the Photon Factory (Cho et al. 1988; Hirata et al. 1990) have been made with high incident fluxes and consequently with the MCP acting as a current amplifier. Mapping these quantum efficiencies obtained in current mode onto the desired pulse counting efficiency would be a far from straightforward task, needing to take into account MCP current limitation (Fraser et al. 1991a; Fraser, Pain & Lees 1993) and gain lifetime effects (Fraser, Pearson & Lees 1988). Even
if this were possible for uncoated MCPs, exposing a CaI or KBr coated plate to synchrotron fluxes of order $10^{11}$ photons per second would lead to the radiation damage problems detailed in Chapters 8 and 9. The energy tunability of the synchrotron source is thus only useful for MCP calibration if the beam intensity can be heavily curtailed. Section 6.3 describes an experiment undertaken at the Daresbury SRS with very significantly reduced flux. Sections 6.4 and 6.5 then discuss the original data obtained and the calibration of the beamline respectively. Section 6.6 examines the absorption edge structure present in the data and finally 6.7 discusses how the lessons learnt in this proof-of-concept experiment, conducted in June 1994, have been used in more recent HRC-S calibration activities. The experiment was conducted by Jim Pearson and John Lees with the assistance of Paul Bailey, the beamline manager; all of whom I thank. The analysis and calibration work is my own based on information from Paul Bailey and the beamline reference manual (Bailey & Quinn 1993).

6.3 Experimental Details

Measurements were conducted on SRS photoemission Beamline 6.1, shown in Figure 1.4 and briefly described in section 1.3.1. The monochromator exit slits were 100 $\mu$m wide, giving an energy resolution of 0.13 eV at 80eV and 0.35eV at 160eV. The absolute energy calibration of the beamline is believed to be correct to better than 0.5eV, at least for normal "high ring current" operation.

The largest part of the flux reduction was achieved by decreasing the number of orbiting electrons in the synchrotron ring. The orbiting current was reduced from 200mA to $\approx 1.2\mu$A; i.e. by a factor of almost $2\times10^5$. This "low ring current" operation mode has now been used on four occasions and has proven consistently reproducible with a stable orbiting current (Owens et al. 1995).

Testing on the peak of the beamline response showed even this reduction not to be enough, to avoid the effects of MCP gain suppression. A further reduction in flux was achieved by vertical steering of the electrons out of their normal orbital plane. The beamline flux is thus extracted from the edge of the orbiting electron distribution rather than the centre, leading to a flux reduction by a factor of approximately twenty.
Operation in low ring current meant that no suitable reference counter was available that could operate at such low fluxes. We would therefore have to rely on the beam calibration already carried out by Daresbury staff. Unfortunately, this calibration data were only available for normal operation mode (i.e. high ring current) and worse still was known to be altered in a complex way by steering the beam out of plane. These problems and those associated with second order grating contamination (as no passband filters available either) are addressed in Section 6.5.1

The experimental configuration inside the experimental chamber is shown in figure 6.3. The microchannel plates used were uncoated 36mm Mullard (now Philips Photonics) devices dating from the mid-nineteen-eighties ROSAT HRI programme. Both plates had a 12.5 μm pore size on an interchannel pitch of 15 μm yielding an open area fraction of 63% and a pore length-to-diameter ratio of 80:1. The plates were arranged in the usual chevron configuration with the front plate having a 0° bias angle and the rear plate 13°. An applied voltage of 1150V was maintained across each plate and 200V potential difference between the rear plate and a steel non-imaging anode used as the electron pulse readout. No facility existed to apply an electron accelerating potential between the rear of the front plate and the front of the rear plate as would occur in the optimum AXAF HRC configuration. The whole assembly, mounted on a manipulator, could be accurately rotated to incident angles of 10° and 40° with respect to the synchrotron beam. The detector and vacuum system were vacuum baked for 10 hours at 150 °C before measurements began.

6.4 Results

The description above is of a replacement pair of MCPs, the first pair (with a solid glass rim) having cracked during vacuum bake. Under time pressure (the measurements described here were all conducted in our last six hours of beamtime) the replacement MCPs had to be operated whilst not fully cooled following vacuum baking. This factor, coupled with the poor quality of the plates led to unusually poor noise characteristics which could only be countered by operation at lower-than-ideal voltages across each plate. The absence of a potential difference between the plates and low voltages across each plate would be expected to produce broad pulse height
ELECTRODES MAINTAINING A 1150V POTENTIAL ACROSS EACH PLATE

CENTRING RIG

FRONT GROUND PLANE MESH
FRONT MCP $0^\circ$ BIAS
REAR MCP $13^\circ$ BIAS

STAINLESS STEEL ANODE (non imaging)

to preamp

Figure 8.3: MCP calibration assembly rotated at 10 or 40 degrees to the beam
Figure 6.4: A typical pulse height distribution (PHD) obtained from the MCP detector.

distributions (PHDs) which as demonstrated by the sample PHD of figure 6.4, turned out to be the case.

Under the described conditions the noise count rate above a fixed 0.5pC lower level charge discriminator was of order 40 ct s\(^{-1}\) compared to a signal count rate of order 1000 ct s\(^{-1}\). All the data presented here has been noise subtracted.

Figure 6.5a shows raw data collected at an angle of incidence of 40°. The dashed vertical lines represent absorption edges of the atomic constituents of Philips' standard type 3502 MCP lead oxide glass and of the nichrome (Ni\(_7\)Cr\(_2\)Fe\(_3\)) electrode coating (Fraser et al. 1982). The shape of the curve mainly corresponds to the response of the monochromator, which has not been calibrated out in this figure. Figure 6.5b shows higher resolution data (E/\(\Delta E\) = 1000 as opposed to 200) at the same angle of incidence (40°), over the energy range 95 to 130 eV where the most significant structures in a) are visible. These measurements show the level of detail obtainable with data taken in 0.1eV steps.

Figure 6.6 illustrates the data obtained at an angle of incidence of 10°. The increase in count
rate corresponds to the well known increase in MCP efficiency with decreasing grazing angle of incidence (Fraser et al. 1984; Fraser 1992). A comparison of the data at both energies along with analysis of possible physical interpretations of the structures seen is made in Section 6.6.

First, in section 6.5, a detailed description of the analytical process used to normalise the data for source, mirror and monochromator responses is presented.

6.5 Calibration of the Beamline Response

The starting point for data normalisation is the beamline flux versus monochromator energy response for the operational mode used (range 2, recoated blazed grating). This data was obtained from Paul Bailey, the beamline manager and is displayed graphically in the beamline 6.1 reference manual (Bailey & Quinn 1993) and figure 6.7 below. Note this data was taken in normal high beam current mode; no calibration data at low beam currents is available.

The output curve of figure 6.7 was obtained from normal incidence drain current measurements from a clean copper sample without any second order filter, and then corrected for the Cu efficiency at the primary energy from the data of Day et al. (1981). There will, however, exist a significant fraction of the flux at the second order (grating n=2) energy. The assumed Cu efficiency, therefore needs to be corrected for the second order content as the reference photocathode efficiency at the second order energy will be different.

6.5.1 Second Order Contamination

Values for the second order contributions were again obtained from the beamline reference manual (Appendix A.10). Values taken are those for range 2 with the recoated blazed grating. These measurements were made by placing a Cu sample in the beam and measuring the energy spectrum of the emitted electrons. The values for fractional second order content are then calculated from the relative strength of the electron spectrum peaks below the primary and second order energies. The peak in the 3D Cu photoemission band about 5eV below the Fermi edge for photons of the required energy is used for this purpose. i.e. for 80 eV photons the Fermi edge is at an electron energy of approximately 75 eV and the height of the peak in the
Figure 6.5: (upper) Noise corrected MCP count rate data at an angle of incidence of 40°; (lower) repeat measurement, with 0.1 eV resolution at 40° over the restricted energy range 95 to 130 eV where the most significant structures were visible.
Figure 6.6: MCP count rate in the 75 to 200 eV band. Angle of incidence 10°

Figure 6.7: Flux versus energy profile for beamline 6.1 in normal high ring current mode. Refers to a ring current of 200mA
electron spectrum at 70 eV is measured. The electron spectrum peak at 150 eV due to second order photons was also measured and the second order fraction calculated from,

\[
s_c = \frac{Y(2E-10)}{Y(2E-10) + Y(E-10)}
\]  \hspace{1cm} (6.1)

where \(Y(2E-10)\) is the height of the electron spectrum peak at approximately 10 eV below twice the photon energy, \(E\), and \(s_c\) is the calculated second order fraction.

This calculation, however, takes no account of the copper efficiency differences at both energies. The true second order fraction is thus;

\[
s = \frac{Y(2E-10)/\kappa(2E)}{Y(2E-10)/\kappa(2E) + Y(E-10)/\kappa(E)}
\]  \hspace{1cm} (6.2)

where \(\kappa(E)\) is the photoionisation cross-section for the Cu 3D band from the data of Yeh & Lindau (1985). Rearranging equation 6.1 for \(Y(2E-10)\), then substituting into equation 6.2, multiplying top and bottom by \(\kappa(2E)\) and dividing top and bottom by \(Y(E-10)\) yields a method of calculating the true second order fraction from the recorded \(s_c\) values.

\[
s = \frac{1-s_c}{\kappa(2E)} \cdot \frac{1-s_c}{1+s_c + \kappa(2E)/\kappa(E)}
\]  \hspace{1cm} (6.3)

Values of the second order fraction thus calculated are presented in table 6.1 together with the generally lower uncorrected values presented in the beamline reference manual.

**Correcting the Beam Profile**

Knowing the second order content, the flux versus energy profile of the beam can be corrected for second order content to obtain the real flux levels at each energy. First, the copper efficiencies at both the primary and secondary energies were calculated from the data of Day et al. (1981)
Table 6.1: Values of the second order content of beamline 6.1 as a function of energy.

<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>Second Order Fraction ($s_2$) (Bailey &amp; Quinn 1993)</th>
<th>True Second Order Fraction ($s$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>40</td>
<td>0.293</td>
<td>0.334</td>
</tr>
<tr>
<td>50</td>
<td>0.166</td>
<td>0.206</td>
</tr>
<tr>
<td>60</td>
<td>0.140</td>
<td>0.221</td>
</tr>
<tr>
<td>70</td>
<td>0.086</td>
<td>0.156</td>
</tr>
<tr>
<td>80</td>
<td>0.046</td>
<td>0.089</td>
</tr>
<tr>
<td>90</td>
<td>0.025</td>
<td>0.058</td>
</tr>
<tr>
<td>100</td>
<td>0.006</td>
<td>0.017</td>
</tr>
<tr>
<td>120</td>
<td>0.004</td>
<td>0.003</td>
</tr>
</tbody>
</table>

assuming a straight line fit between data points. The real efficiency of the Cu to the combined 1st and 2nd order contributions of the beam could then be calculated by multiplying the efficiencies by the relative contributions of the two energies.

In mathematical representation:

$$\Psi_{Cu} = s \chi_{Cu}(2E) + [1 - s] \chi_{Cu}(E)$$  \hspace{1cm} (6.4)

where $s$ is the true second order fraction, $E$ the primary photon energy $\Psi_{Cu}$ the effective copper efficiency to both primary and second order contributions to the beam and $\chi_{Cu}$ the copper efficiencies of Day et al. (1981). With the photocurrent as measured by the copper reference counter denoted $I(E)$ and the beam flux denoted $F(E)$, both at monochromator setting energy $E$ we have;

$$I(E) = \Psi_{Cu}.F(E)$$  \hspace{1cm} (6.5)

or substituting for $\Psi_{Cu}$ ;

$$I(E) = (\chi_{Cu}(E).[1 - s] + \chi_{Cu}(2E).s).F(E)$$ \hspace{1cm} (6.6)

The flux versus energy beam profile of figure 6.7, now denoted $F_{set}(E)$ can then be related to the true flux, $F(E)$, by ;
The total flux versus energy curve, \( F(E) \), can then be split into its primary and secondary components using the second order fraction in the beam calculated previously.

\[
F_{\text{est}} = \frac{F(E)}{\chi_{\text{Cu}}(E)} = \left( \frac{[1 - s] \chi_{\text{Cu}}(E) + s \chi_{\text{Cu}}(2E)}{\chi_{\text{Cu}}(E)} \right) F(E) \quad (6.7)
\]

The total flux versus energy curve, \( F(E) \), can then be split into its primary and secondary components using the second order fraction in the beam calculated previously.

\[
\text{Primary Flux}(E) = [1 - s] F(E) = [1 - s] \left( \frac{\chi(E)}{[1 - s] \chi(E) + s \chi(2E)} \right) F_{\text{est}} \quad (6.8)
\]

\[
\text{Second Order Flux}(E) = s F(E) = s \left( \frac{\chi(E)}{[1 - s] \chi(E) + s \chi(2E)} \right) F_{\text{est}} \quad (6.9)
\]

**Calculating the true MCP efficiency**

Knowing the total flux at both the primary and second order energies for all monochromator settings is only the first step. What we would like to find is the MCP efficiency versus primary energy with all effects of the second order photons removed. Let \( N \) be the noise subtracted MCP detector count rate and \( Q \) the MCP quantum efficiency in counts per photon. Then

\[
N = [1 - s] F(E) Q(E) + s F(E) Q(2E) \quad (6.10)
\]

therefore

\[
Q(E) = \frac{N}{[1 - s] F(E)} - \left( \frac{s}{1 - s} \right) Q(2E) \quad (6.11)
\]

To solve equation 6.11 for \( Q(E) \) we also need to know the MCP efficiency to photons at all the second order energies, \( Q(2E) \) - which is essentially the same problem as finding \( Q(E) \). We know, however, that the second order contribution decays away with increasing energy (Table 6.1) and is so small above a certain energy (\( \sim 120 \text{eV} \)) as to be negligible. Table 6.1 shows a second order content below \( \frac{1}{2}\% \) at 120 eV so it seems reasonable to take this as the cut off energy. Thus, above 120 eV the MCP quantum efficiency is solely due to 1st order photons:
These higher energy MCP efficiency values can then be used as second order efficiencies $Q(2E)$ to correct the data at half the original energy. In this way, if the data set covers a wide enough range of energies, all the second order contribution can be removed.

Obviously the normalisation works best for a large energy range data set, so three of the 40° angle of incidence data sets were combined to create a set covering the range 40 to 300 eV. This unnormalised data is presented in figure 6.8. Note that it contains the 75 to 200 eV data from figure 6.5a. Figure 6.9 shows the quantum efficiency of the MCP at 40° without correction for second order effects, i.e. it is simply the outcome of dividing the counts per second data with the expected flux at each energy. The data of figure 6.7 divided by 170,000 is used as the expected flux. The factor of 170,000 arises from the reduction in electron ring current from 200mA to 1.2μA. The effect of beam steering has been accounted for by multiplying the efficiencies by a factor of 20, to produce our best estimation of the absolute efficiency. The flux reduction due to beam steering is poorly known, so the efficiencies presented on the following graphs should only be taken as being correct to within a factor of two.

Figure 6.10 displays MCP efficiency data obtained, using the second order correction procedures outlined above, for the 40° grazing incidence data set. Figure 6.11 shows the similar plot for 10° grazing incidence. The limited energy range over which measurements were conducted at 10° means that second order contamination is of less concern.

Comparable experimental and theoretical efficiencies are also presented in figure 6.11, while the synchrotron measurements of Hirata et al. (1990) are displayed in Figure 6.12. Each of these measurements were made on bare MCPs, at 8° (Kowalski et al. 1986), 10° (present data and Fraser (1982) model) or 13° (Hirata et al. 1990) grazing incidence. The Hirata et al. (1990) measurements are of the current yield; that is the output current from the MCP is recorded, whereas the other experiments counted the number of electron pulses detected. Pulse counting is the normal mode of operation for low count rate detectors (such as those used in astronomy). A current yield calibration would likely enhance the efficiency at higher photon energies relative to low energies when compared to a pulse counting calibration because more electrons would be

\[
Q(E \geq 120\text{eV}) = \frac{N}{F(E)} \quad \text{as} \quad s \sim 0
\]
Figure 6.8: Combination of three unnormalised data sets at 40° grazing angle of incidence.

Figure 6.9: Quantum efficiency for a MCP at 40° grazing incidence, uncorrected for 2nd order effects
Figure 6.10: Relative quantum efficiency for a MCP at 40° grazing incidence, corrected for the second order contribution to the beam.

Figure 6.11: Relative quantum efficiency for a MCP at 10° grazing incidence, corrected for the second order contribution to the beam. ○) Experimental data of Kowalski et al. (1986) at 8° grazing incidence; △) Theoretical predictions of uncoated MCP efficiencies at 10° grazing incidence by Fraser (1982).
Figure 6.12: 60 to 600 eV MCP current response of Hirata et al. (1990) with a 13° angle of incidence.

Agreement between the four data sets is severely limited, with relative wavelength independence between 120 and 200eV being the only similarity. The data of Hirata et al. (1990) shows a significant decrease in efficiency above approximately 160eV, but this may be due to carbon contamination evident from the carbon K edge at 284.2 eV. The absolute efficiencies from the synchrotron measurements of Kowalski et al. (1986) also agree well with the predictions of Fraser (1982) in this energy region, especially when the difference in angle of incidence is taken into account. The efficiencies from synchrotron measurements presented here differ by a factor of 10, which is far greater than the beam steering. Below 120eV the data of Hirata et al. (1990) and Kowalski et al. (1986) see a major reduction in efficiency, while the predictions of Fraser (1982) remain flat.

A possible explanation for the difference between our measurements and the others is that the flux versus energy profile used to calibrate our data was not consistent with the true flux response of the beamline. This is at least possible as the profile has never been measured for low ring current operation. A test of the credibility of the "beam versus energy" hypothesis is to take our uncalibrated data and divide it by MCP efficiency data which should yield the true flux response of the beamline. The isolated energy data of Kowalski et al. (1986) was selected for this purpose. The beam response thus generated is shown in the graph. 

Agreement between the four data sets is severely limited, with relative wavelength independence between 120 and 200eV being the only similarity. The data of Hirata et al. (1990) shows a significant decrease in efficiency above approximately 160eV, but this may be due to carbon contamination evident from the carbon K edge at 284.2 eV. The absolute efficiencies from the synchrotron measurements of Kowalski et al. (1986) also agree well with the predictions of Fraser (1982) in this energy region, especially when the difference in angle of incidence is taken into account. The efficiencies from synchrotron measurements presented here differ by a factor of 10, which is far greater than the expected errors caused by beam steering. Below 120eV the data of Hirata et al. (1990) and Kowalski et al. (1986) see a major reduction in efficiency, while the predictions of Fraser (1982) remain flat. On the other hand the synchrotron measurements record a rapidly rising efficiency at low energies which is totally unexpected.
Figure 6.13: Crosses- Flux response of beamline 6.1 at full ring current and zero vertical beam steering. Full curve- result of dividing MCP counts per second data at $40^\circ$ with the expected MCP quantum efficiency of Kowalski et al. (1986).

in figure 6.13 together with the high ring current measurement. The difference in energy of the peak in the flux response can be seen to be approximately 50eV. The structure present in the calculated curve are a result of absorption edge effects which cannot be calibrated out by using line energy data.

The possible cause of such a discrepancy is, however, not obvious. Steering the electron beam off its normal path would produce a tiny deviation in the angle at which the X-ray beam strikes the beamline optics. The lower section of figure 1.4 shows the vertical positions of the various beamline optics; from this figure it can be seen a vertical shift in the source position will not affect the pre-mirror, but will change the angle at which the X-ray beam meets the monochromator grating. The change in the angle of incidence, $\delta \theta$, depends on the source-to-monochromator entrance slit distance as,

$$
\tan \delta \theta = \frac{\text{vertical steering}}{\text{source - slit distance}} = \frac{0.0028}{9.126} \quad (6.13)
$$

with distances in meters. Thus $\delta \theta = 0.018^\circ$ compared to the grazing angle of incidence of $\sim 7^\circ$. This would cause a 1.5eV shift in the energy passed by the monochromator for a 154eV setting,
but would not significantly alter the flux versus energy profile. The reduction in the number of orbital electrons could also not be expected to effect their emission distribution. It has indeed been shown by later experiment that normalisation of low current data by the high current beam response produces believable data (Pearce et al. (1995) and Figure 6.14).

Another possibility is that in high ring current mode the intense throughput warps, through heating, the precollimator optics in a way which is absent in our experiment. Probably the most plausible explanation though is that the effects of the poor MCP quality, possibly coupled with gain suppression effects and low operating voltage, had radically altered the MCP efficiency response. Count rates of one to two thousand counts per second were concentrated in an area of approximately 2 square millimeters (Figure 1.5) corresponding to ~ 0.1 counts per channel per second. For reasonable quality MCPs, this would only cause a minor reduction in efficiency (Fraser et al. 1991a). In our case however the poor PHD (Figure 6.4) indicates that the real number of counts through the detector is far in excess of the counts above the lower level discriminator and this could cause a significant reduction in efficiency toward the beamline response peak. This cannot wholly explain the apparent shift of figure 6.13 as a second peak the other side of the beamline peak would be expected. Little is known of how poor pulse height distributions, high temperatures and low operating voltages would effect gain suppression effects so I suspect a full explanation of these measurements will never be made.

What can be said, however, is that our lack of understanding of the quantum efficiency measurements does not inhibit an analysis of the interesting absorption edge structure seen in figures 6.5 and 6.6.

### 6.6 Physical Interpretation of the Edge Structures

Taking first the data at 40° grazing incidence data (figure 6.5), a number of significant structures are evident, not all of which appear correlated with a marked edge. It should be noted though that the position of edges is affected by neighbouring atoms, the vertical line positions in the figure referring to pure elements. Figure 6.5b plots the uncalibrated MCP quantum efficiency over the energy range where the most significant structures appeared earlier. This data, taken in 0.1 eV monochromator steps, shows the three major features, at 108eV, just above the Pb O_{II}
edge, at 115eV and 125eV which seem uncorrelated with any edge. What can be said though is that the 108, 115 and 125 eV features do not correspond to the Silicon L\textsubscript{II} or L\textsubscript{III} edges. In fact the photoemission maximum for silicon oxide systems is predicted to occur at 104eV (Bell & Ley 1988). Instead the 108eV feature at least, appears to be due to lead in the glass (Pb O\textsubscript{II} edge). The Pb O\textsubscript{III} edge at 84eV also has an associated bump. That the major features are lead induced is supported by the data at 10 degrees grazing incidence. This, shown in figure 6.6, has less prominent features, with the 108eV peak especially reduced. This would be expected, if the features were due to lead, because at 10 degrees the photons penetrate less deeply into the channel walls and within 100Å of the surface, lead is known to be removed as a consequence of the hydrogen reduction process used during microchannel plate manufacture (Wiza 1979).

There is however a complication to this conclusion. The pulse height distributions (PHDs) were compromised by below par MCP quality and lack of an inter-plate potential. For a grazing incidence angle of 40° the PHDs were poor, as demonstrated by figure 6.4, but for 10° grazing incidence they were much worse. Poorer PHD quality could lead to a smoothing of spectral features as a result of electrons emitted just above the absorption edge being of low energy and more likely rejected as background noise. It certainly appears that even ignoring the 108eV edge, figure 6.6 is far smoother than the corresponding 40° plot (figure 6.5).

Comparing the data presented here with that of Hirata et al. (1990) (figure 6.12), is also interesting. The differing shapes of the response curves are obviously due to the inability to correctly calibrate our data. In terms of absorption edges similar structures around 110eV are visible, but the most striking features are the oxygen K edge (at 543 eV, out of the range of our data) and the carbon K edge at 284eV which isn’t present in our data of figure 6.8. Carbon is not a constituent of MCP glass, but is a possible contaminant and would have a large effect if present in anywhere near the concentrations seen by Hirata.

6.7 Conclusions and Recent HRC-S Calibration Activity

Experimentally, the proof of concept calibration attempt described here was a success. We learnt that synchrotron operation with a low ring current (~ 1 μA) was feasible for the calibration of the HRC-S detector. Also the experiment confirmed that sparse calibration of spectroscopic
soft X-ray instruments by using available emission line energies and assuming simple steps at absorption edges would introduce an unacceptably large degree of spurious structure (including line emission and absorption) into future cosmic source spectra. That said, a number of problems requiring future solution were thrown up. Most importantly, the apparent difference in shape of the beam strength versus monochromator energy at normal synchrotron fluxes and using the set up as described above.

We have also shown the quantum efficiency exhibits a complex dependency upon the angle of incidence, especially where the X-ray penetrates to different channel thicknesses and thus different lead concentrations in the glass. This would require future calibrations to be conducted at the angles of incidence to be used during the AXAF mission.

In January 1995 a second calibration experiment was conducted on KBr and CaI coated microchannel plates using beamlines 4.2 (between 2 and 6 keV) and 6.1 as reported by Pearce et al. (1995). With the cause of the data normalisation problems unknown every effort was made to ensure possible causes were eliminated. Most importantly, a method of applying an inter-plate potential was used; this and the better quality of the MCPs improved the pulse height distributions and allowed accurate measurements to be made at lower grazing angles. A further reduction, of a factor of two, in ring current removed the requirement to use beam steering to reduce the flux. These changes and the analysis procedure developed above led to the production of normalised data comparable to line energy measurements, but with the fine structure only synchrotron calibration could produce. Figure 6.14 shows the result of the January 1995 calibration on a bare MCP at 14° angle of incidence compared to the line energy measurements of Kowalski et al. (1986).

Further measurements were conducted during January 1996, between 300eV and 2keV to complete the operational range of the HRC-S detector.
Figure 6.14: Comparison of bare MCP quantum efficiency synchrotron calibration [*'s courtesy of S. E. Pearce] with the line energy measurements of Kowalski et al. (1986) [full curve].
Chapter 7

Quantum Efficiency Measurements on Alkali Halide Photocathodes

7.1 Introduction

Caesium Iodide in particular and the alkali halides in general are well known to be highly efficient photoconverters at soft X-ray and extreme ultraviolet wavelengths. Of the alkali halides CsI generally has the largest quantum efficiency, that is the ratio of escaping photoelectrons to impinging photons. This is a result of its exceptionally large electron escape length (Akkerman et al. (1994) found that a 1eV electron had an escape length of order 16nm) and its low electron affinity ($E_a = 0.1 - 0.2$ eV). Caesium Iodide photocathodes have thus found use in Parallel-Plate and Solid Photocathode Avalanche Chambers (PPAC and SPAC) (Charpak et al. 1993), as readouts for Ring Imaging Cherenkov (RICH) detectors (Lu et al. 1995) and gas scintillation detectors (Dangendorf et al. 1990), and as channel coatings in Microchannel Plate (MCP) detectors (Fraser et al. 1982).

For the purposes of calibrating any of these detectors and comparing the alkali halide photoconverter to gaseous detectors, accurate quantum efficiency measurements over a wide energy range are required. This has long proven a significant problem, especially at far UV where measurements by different groups yielded quantum efficiencies varying by a factor of 3. A good review
of these UV discrepancies is given by Breskin (1996), which concluded that most inconsistencies had been clarified by a better understanding of thin film preparation and characterisation procedures, surface phenomena and the effects of electric fields. At X-ray energies surface effects, which seem most important in the UV measurements, are less important and although there is an apparently rogue measurement by Alexandropoulous (1976), X-ray quantum efficiency determinations by Eliseenko, Shchemelev & Rumsh (1968), Day et al. (1981) and Henke, Knaser & Premaratne (1981) are all in good agreement.

The measurements presented below do not represent the absolute quantum efficiency of ‘fresh’ photocathodes as they were obtained from highly radiation damaged alkali halides. Where the data is useful, however, is in providing a continuous energy calibration. We may scale the synchrotron measurements to account for radiation damage effects so they match accepted measurements conducted at a number of X-ray line energies hence obtaining a believable broad band efficiency calibration, continuous in energy. Efficiency measurements were also obtained at a wide range of grazing angles.

7.2 General Experimental Description

The quantum efficiency measurements presented here are simply a useful by-product of two other research topics described in this thesis; namely the investigations of the X-ray vectorial effect presented in chapters 2 to 5 and alkali halide radiation damage presented in chapters 8 and 9. Detailed descriptions of the experimental configurations used to produce the data presented below can thus be found in section 3.1 for the polarimetry run on beamline 6.3 (section 1.3.2, energy 3-6 keV) and section 8.2 for the radiation damage run on beamline 3.4 (section 1.3.3, energy 2-4 keV). Alkali halide photocathodes were mounted under vacuum on a drive, capable of accurate rotation to a series of grazing incidence angles. The sample was then irradiated at a fixed monochromator energy for between 10 and 20 minutes to escape the region of rapid radiation degradation. By variation of the monochromator setting the photon energy was then changed while the drain current from the photocathode and a reference foil monitor in front of the sample were recorded. The flux reaching the photocathode, \( F \), is then the current from the reference foil divided by its efficiency at the monochromator energy all multiplied by
the foil transmission. Aluminium and beryllium foils were used as reference counters; their transmission properties were derived from Zombeck (1980) and their quantum efficiencies from Alexandropoulos (1976). The photocathode efficiency, in electrons per photon, is then simply the drain current in electrons divided by $F$. For the polarisation sensitivity measurements of chapters 2 - 5, the average quantum efficiency of the measurements conducted in both photon polarisation states was used. During the radiation damage examinations, all measurements were conducted at normal incidence.

7.3 Measurements

Figures 7.1 and 7.2 show caesium iodide photoyield data between 3 and 4 keV obtained during the polarimetry experiments described in chapter 2. The steady increase in quantum efficiency approaching grazing incidence, as expected from geometric factors, shows that radiation damage has been of a similar magnitude during each measurement. This is less true of the data in the 2 to 3 keV band, where as illustrated in figure 7.3, efficiencies at 10° and 12° grazing incidence yield similar values. The overlap between the two energy ranges at 3000eV does however show reasonable agreement. During a 2-3keV scan the 3000eV monochromator setting is reached after one hour’s irradiation, whereas coverage of the 3-4keV range corresponds to only 15 minutes irradiation. This shows that the severity of CsI radiation damage isn’t too large in the period during which measurements were made.

The final CsI photoyield measurement on beamline 6.3 was conducted over the energy range containing the L absorption edges of both caesium and iodine and is displayed in figure 7.4. The inset displays this same data taken at 8° grazing incidence alongside lower energy data taken at the same angle of incidence. The 3eV energy resolution enables significant structure above the absorption edges to be seen. X-ray Absorption Fine Structure (XAFS), the oscillatory variation of photoyield extending over 100 eV above the edge is visible on all edges and results from interference of outgoing and scattered (from neighbouring atoms) photoelectric waves in the region near the nucleus of the absorbing atom. White lines, (sharp peaks in efficiency on the absorption edge) can be seen on Cs $L_{II}$ and $L_{III}$ edges. White lines occur when the electron can be promoted to a partially filled band with complex structure due to perturbations
Figure 7.1: The variation of Caesium Iodide photoyield with energy for five grazing angles of incidence; namely 6° (+), 8° (x), 10° (o), 12° (□) and 15° (△).

Figure 7.2: Caesium Iodide photoyield versus photon energy at 20° (+), 30° (x) and 60° (□) grazing incidence and normal incidence (o).
from neighbouring atoms (Shah & Gurman 1982). In Caesium the 6s level is unfilled and the transition from the 2p state, corresponding to the L_{II} and L_{III} edges, to the 6s level is allowed by quantum mechanic consideration, i.e. a change by 1 in the angular momentum number, so we see white lines. For Iodine it is the 5p shell that is unfilled, so transition from the 2p band is forbidden and no white lines appear on the L_{II} or L_{III} edges, but should on the L_{I} edge at least if the 5p band is sharply peaked in density of states terms. An interesting comparison to our data is provided by the linear absorption coefficient data for crystalline CsI shown in figure 7.5. The similarity in structure shows photoemission from our samples has not been greatly compromised by radiation damage or our procedure for evaporating CsI onto the substrates.

Alkali halide photoyield data was also obtained on beamline 3.4 during the radiation damage measurements described in chapter 8. Figure 7.6 shows the normal incidence photoyield measurements on both pure caesium iodide and thallium doped CsI. Thallium doping is commonly used to enhance the scintillation light output of CsI and to increase the wavelength of emission improving its subsequent detection by a photomultiplier tube. In this case, however, the Tl doped sample suffered greater radiation damage during the pre-measurement period leading to
Figure 7.4: Caesium iodide photoyield at 8° grazing incidence across two energy ranges covering the region of the L edges of both Cs and I. Inset: a combination of the CsI photoyield data obtained at 8° grazing incidence showing the good correspondence between measurements over different energy ranges.
Figure 7.5: Relative linear absorption coefficient versus photon energy across the L absorption edges of caesium and iodine (Haelbich, Iwan & Koch 1977).
the difference in quantum efficiency seen in figure 7.6. The five M absorption edges of thallium in this energy range can be seen to have no effect on the quantum efficiency, which is hardly surprising given the thallium is only present on 0.1% of the anion sites. The pure CsI photoyield data of Day et al. (1981) is also plotted in figure 7.6, in comparison our data shows a 40% higher efficiency from what is already a radiation damaged sample (15 minutes irradiation before commencing measurements led to an approximate efficiency reduction of 10%). Measurements on beamline 6.3 gave a third distinct value for the normal incidence photoyield (~ 10% at 3300 eV c.f. 42% (Day et al. 1981) and 60% (beamline 3.4)) indicative of the problems obtaining absolute photoyield calibrations when radiation damage is so prevalent. The agreement in the shapes of the three curves in figure 7.6 and the beamline 6.3 measurements presented earlier in this chapter is, however, excellent.

A photoyield measurement on potassium bromide was also made on beamline 3.4 where, as shown in figure 7.7, two absorption edges are prominent. This measurement also conducted at normal incidence shows the quantum efficiency of KBr to be only a quarter of that for CsI, although radiation damage and the other effects discussed above make absolute comparison difficult.

### 7.4 Summary: Quantum Efficiency Determinations

The level of detail in MCP and photocathode quantum efficiency responses seen in these two chapters demonstrate why it is vital to use broadband techniques to calibrate detectors with 1eV resolution. For this the Daresbury synchrotron in low ring current is an ideal source, but the difference between normal and low ring current, especially the prospect of heat from a normal white beam warping the pre-monochromator optics, requires the synchrotron beamlines to be themselves first calibrated by a detector with a known response such as a proportional counter. Even then, in the absence of reference counters, scaling the broadband response obtained with more precisely known line energy efficiencies remains best practice.
Figure 7.6: Normal incidence photoyield of pure (+) and thallium doped (x) caesium iodide compared to the data of Day et al. (1981) for pure CsI. The present data represents the response of a heavily radiation damaged sample.

Figure 7.7: The quantum efficiency of Potassium Bromide in the region up to the potassium K edge at normal incidence.
Chapter 8

Radiation Damage in Alkali Halide Detectors

8.1 Introduction

Alkali halide photocathodes have been shown to be highly efficient X-ray detectors. For example, results published in the previous chapter show CsI has a normal incidence photoconversion (from X-rays to electrons) efficiency of unity at 2500 eV, compared to less than 10% for metals (Henke, Knauer & Premaratne 1981; Day et al. 1981; Fraser et al. 1992). Caesium iodide is also a highly efficient X-ray scintillator material, although mainly doped with thallium or sodium to further increase light yield and increase the wavelength of emission such that it is more efficiently detected by photomultiplier tubes (PMTs). In both applications, the most significant problem in using CsI or other alkali halides in reasonable high flux environments is that of radiation damage. It has been demonstrated that (a) during periods of X-ray exposure, there are significant reductions in the photoyield (Breskin 1996) and scintillation yield (Jing et al. 1996) from CsI detectors and (b) after a break in exposure the photoyield rapidly recovers (Woody et al. 1992). Both these effects make the comparison of results from alkali halide detectors taken at different times difficult, as well as introducing the possibility of having to regularly replace the photocathode in some applications.
The aims of the research reported in the subsequent two chapters are to characterise variations in CsI and KBr quantum efficiency under X-ray bombardment and to examine possible ways of reducing damage. The remainder of this chapter describes the experimental programme while Chapter 9 describes a model which provides insight into how processes at the atomic level lead to the quantum efficiency reductions witnessed. The majority of research to date has been on the degradation of the photoconversion efficiency although scintillation decay is addressed in sections 8.4 and 9.10.

8.2 Experimental Details

Initial radiation damage measurements were conducted on beamline 6.3 at the Daresbury SRS immediately following the polarimetry measurements described in Chapter 3. Details of the experimental configuration, chamber alignment procedure and of the in situ preparation of the CsI samples can be found in section 3.1. The General characteristics of beamline 6.3 are described in section 1.3.2 and in more detail in the beamline reference manual (MacDowell, Norman & West 1986).

Measurements were conducted at three grazing angles of incidence, (10°, 25° and 40°) in both beam polarisation states and at two photon energies, 2500eV and 3500eV. A single coated sample, divided into regions as illustrated by figure 8.1, was used to save time. Each measurement could be conducted on a fresh area of CsI. The following sequence was adopted for each measurement;

1. The beam was blocked from the experimental chamber by means of one of the gate valves along the beamline.
2. If necessary the monochromator was then tuned to give the required photon energy.
3. The sample was then moved so that the beam would fall onto the required spot and at the correct angle of incidence and beam polarisation. The drive on which the sample was mounted had two rotational and one linear degree of freedom.
4. Recording of the values of drain current from the sample and reference counter was begun;
only then were the beamline stops removed and the sample illuminated.

5. After 30 minutes the beam was again blocked, while the recording of measurements continued. Then after a further 30 minutes the valves to the beam were reopened for another 30 minutes irradiation. The "30 minutes out, 30 in" cycle was then repeated once more to examine the nature of the recovery of quantum efficiency. The valves were then finally shut to allow for the setting up of the next measurement.

In October 1994, almost two years after the beamline 6.3 measurements, a generous colleague, Dr A. Owens allowed me 24 hours of his beamtime to conduct a follow up study. These measurements were conducted on beamline 3.4 (described in section 1.3.3). Samples of CsI, KBr and thallium doped CsI coated onto stainless steel substrates were prepared in Leicester by vacuum deposition and transported to Daresbury under a dry nitrogen atmosphere. The beamline 3.4 vacuum chamber had a linear drive onto which 10 samples were transferred, and pumped down to an operational pressure of order $10^{-8}$ mbar. The experimental procedure was virtually identical to that described above, except that each measurement was on a fresh sample and all measurements were conducted at normal incidence.

### 8.3 Results

Knowing the flux entering the chamber, using a 0.75μm Al foil reference counter, we were able to calculate both the absolute efficiency of the CsI sample and the total flux that had been incident on it. The data is thus plotted in terms of photoyield versus accumulated dose rather than time to allow proper comparison to be made between data taken at different times of day, as the synchrotron ring current decreases, and at different energies where the flux passing through the beamline optics will differ.

Figures 8.2 to 8.5 illustrate the common features of the dozen data sets obtained. Each set showed a marked "two component" signature in the quantum efficiency degradation. At first there is a period of very rapid decay (in figure 8.2 the quantum efficiency is down 20% in the first 5 minutes), followed by a slow decay continuing past 90 minutes. After 90 minutes of irradiation the efficiency of most samples had decreased by approximately half. The data
Figure 8.1: Arrangement of regions on the CsI photocathode for examining radiation damage, so that each measurement can be made on a fresh region of the sample. The spots represent the central beam position of the various known drive settings. The left half of the photocathode was used for measurements with an S polarised beam, and the right side for P polarised measurement. The longest shaded regions were used for measurements at 10° grazing incidence, the mid length ones for 25° and the shortest regions for 40° grazing incidence. Measurements at 2.5 keV were conducted on the lighter shaded regions, the darker shaded regions for those conducted at 3.5keV. All distances are in mm.
presented in figure 8.5 is the exception to both these rules as the overall decay is both less pronounced and almost lacks the fast decay component. This is most likely the result of the measurements starting after the beam had been incident on this region of the sample. Various authors have previously witnessed this two component nature of alkali halide radiation damage and have generally ascribed the fast component to surface effects and the slower one to defect accumulation in the bulk (Breskin 1996).

There are also some striking differences between different decay scans, most surprisingly in the recovery characteristics. All samples irradiated with 2500 eV X-rays exhibited recoveries in efficiency while the beam was blocked off, but all those irradiated by 3500 eV X-rays on beamline 6.3 did not. The cause of such an effect is rather a mystery as conditions during the recovery phase would have been identical, there are no Cs or I absorption edges in the region between 2500 and 3500 eV, the degradation looks similar in all other ways and later measurements conducted on Daresbury beamline 3.4 at 3610 and 3650 eV also exhibited a significant recovery in efficiency. The X-ray absorption length is, however, three times greater at 3500eV than 2500eV so maybe a surface effect, significant only at the high fluxes of beamline 6.3, results in the absence of a recovery. The other curiosity is the variation in the level of accumulated flux over the 90 minute period. Over the six data sets at each energy this ranges from 4.5x10^{13} to 8.5x10^{13} photons at 2500eV and 5.3x10^{13} to 9.2x10^{13} at 3500eV. Such variation is unlikely to be physical, especially as it didn't follow the reduction of the synchrotron ring current, and brings into doubt the accuracy of the reference counter. An inaccurate reference counter would also explain the unusual variation of efficiency with angle of incidence. For instance the 10° grazing incidence data in figure 8.2 has almost the same efficiency as the 40° data of figure 8.3. By simple geometric considerations the absorption depth and hence approximately the efficiency should vary approximately as the cosec of the grazing angle, so a 10° grazing angle should produce a 3.7 times higher photoyield than a 40° grazing angle. The presence of reference counter capable of measurements out by at least a factor of two would at least partly explain the unexpected similarity between observed efficiencies at 10° and 40°. Considering the resulting doubt as to absolute quantum efficiencies and the need for comparability of data, further beamline 6.3 data presented here has been normalised to a relative efficiency of unity after an accumulated flux of 0.2 x 10^{12} photons.

The most significant factor affecting the degradation process was found to be the energy of the
Figure 8.2: CsI quantum efficiency versus 2500eV X-ray dose at 10° grazing incidence. The visible gaps and change of symbol represent the recovery after the sample was allowed 30 minutes out of the beam, before continuing irradiation.

Figure 8.3: Radiation damage by 2500ev X-rays at 40° grazing incidence to CsI.
Figure 8.4: Csl Radiation Damage by 3500eV S polarised X-rays at 25\(^{\circ}\) grazing incidence. Thirty minute periods out of the beam occurred after dosages of \(\sim 35\) and \(67 \times 10^{12}\) here marked by the arrows. No recovery in quantum efficiency occurred during the out of beam periods.

Figure 8.5: Csl Radiation damage by 3500eV P polarised X-rays at 25\(^{\circ}\).
bombarding photons. Figures 8.6 and 8.7 compare the effect of photon energy on the radiation damage process. In both cases it can be seen the speed at which quantum efficiency degradation occurs is faster at 2500 eV than at 3500 eV. Measurements at 1200 eV, displayed in figure 8.8, show that the rate of degradation is even larger at lower energies. Both comparisons suggest photoyield degradation is a result of a near-surface phenomenon, given that the lower the X-ray energy the greater the likelihood of it being absorbed near the photocathode surface. Even though the energy dissipated by a low energy photon is less than a higher energy one, the damage caused near the surface would need to be greater. For measurements conducted at the same energy and angle of incidence, but different polarisation states there is also a visible difference in decay rate. There is, however, no polarisation state for which the decay is consistently less than the other, instead the variation would appear to be random and solely indicative of the experimental errors present.

Quantum efficiency decay measurements taken at the same energy and flux per unit incidence also show a slight angular dependence. Data obtained at 2500 eV and, as displayed in figure 8.9, 3500 eV both show a complex behaviour with measurements at 25° grazing incidence exhibiting the best radiation hardness, followed by 10°, with 40° grazing incidence measurements suffering the greatest radiation damage. The shallower the grazing angle the greater the number of X-ray interactions occur nearer the surface, which as we have seen should lead to greater quantum efficiency degradation. At shallow angles the spot size would also be greater, thus reducing the flux per unit area, which would be expected to have the opposite effect. The weak trend in angular response seen could well be the result of a complex balance between the two effects or more simply be due to experimental errors.

Measurements on beamline 3.4 allowed the effects of other factors in the radiation damage process to be compared. The flux per unit area unfortunately could not be independently altered with other conditions duplicated. Conducting measurements at different photon energies would, however, have the effect of simultaneously changing the rate of flux accumulation. Conducting measurements at the peak and the wings of the monochromator response allowed a wide range of flux rates to be examined. Figure 8.10 shows radiation damage curves for KBr conducted at 2000 eV and 3650 eV for the period of one hour. Although the rate of flux accumulation was 35 times faster at 2000 eV the overall efficiency degradation was only 21% compared to 28% at 3650 eV. This is very hard to explain especially as data presented above suggested higher
Figure 8.6: Comparison of radiation damage scans taken at 10° grazing incidence and 2500eV (+'s and △s - the lower two curves) and 3500eV (the top two curves, □ and o). The different data sets at the same energy and angle of incidence represent different beam polarisation states (top curve S, then P, P and S again).

Figure 8.7: Comparison of decay scans at 2500eV (lower 2 curves) and 3500eV (top curve) for a grazing angle of incidence of 40°. The missing data set exhibited only 20% the efficiency decay seen here indicative that the sample had previously been unintentionally exposed to the beam. Polarisations S, P, S.
Figure 8.8: Degradation of CsI quantum efficiency with 120 eV X-ray dosage. Gaps in the data again represent periods when the beam was shut off for 30 minutes.

Figure 8.9: Comparison of decay scans caused by irradiation by 3500 eV photons at 10° (△'s and +'s), 25° (o and □'s) and 40° (x) grazing incidence.
photon energies caused less efficiency degradation. With the significantly lower flux rate, we may be seeing a very rapid period of degradation which has already occurred in the 30 seconds or so leading up to the recording of the first efficiency value in the peak flux rate experiments. Alternatively the flux could be so large as to completely swamp the radiation damage process or at least a major part of it, say the formation of an alkali metal overlayer \(^1\) on the surface. Calculations presented in the following chapter also show that greater annihilation of defects created in the lattice structure at higher fluxes significantly reduces the dependence on flux rate, at least at these intensities.

Another rather surprising result was obtained by measurements of KBr quantum efficiency decay either side of the potassium K edge at 3608eV. The data sets displayed in figure 8.11 were actually taken at monochromator settings supposedly equivalent to 3610eV and 3650eV as an immediately preceding measurement (figure 7.7) found the edge occurred at a monochromator setting of 3630eV. Interestingly, measurements either side of the K edge exhibit virtually identical reductions in relative efficiency although the linear absorption coefficient is 60% higher \(^2\) above the K edge than below and the absolute efficiency measured was approximately 50% higher.

Finally, figure 8.12 compares radiation damage curves for three photocathode materials at 2000 eV and normal incidence. Rather than plotting all the data points best fit curves to the form of equation 8.1 are shown for clarity. Of the three materials potassium bromide suffered degradation on the fastest timescale although after a long dose accumulation CsI suffered to a roughly similar extent. Thallium doped CsI was degraded on a similar timescale to undoped CsI, but did so to a significantly greater extent.

### 8.4 Scintillation Yield Measurements

Figure 8.13 displays our first measurements of the effect of radiation damage on the scintillation light yield from a Tl doped CsI photovoltaic as measured by the photomultiplier tube \(^3\). Initially

\(^1\) Changes to the crystal structure of the surface and sample bulk during photon irradiation are examined in the following chapter

\(^2\) A description of how the linear absorption coefficients were calculated in presented in section 9.6

\(^3\) Nuclear Enterprises NE-5521
Figure 8.10: Degradation of the quantum efficiency of a KBr photocathode with accumulated flux of 2000eV (+) and 3650eV (△) X-rays. Both measurements were conducted over a period of one hour, but flux accumulation at 3650eV was a factor of 35 slower.

Figure 8.11: Comparison of radiation damage curves for KBr conducted immediately above (△) and below (+) the potassium K edge.
the light yield can be seen to increase before a longer term yield decay sets in. These measurements also conducted on beamline 3.4, show the yield variation with time as opposed to flux. The low rate of flux accumulation at 3610eV explains why it takes longer to reach the peak yield in this case. Given that the damage to the crystalline structure would be identical during both photoelectric yield and scintillation yield measurements it would appear that the physical cause of the fast decay component in photyield causes a rise in scintillation yield, whereas the slow component causes reduced yields in both cases. Curiously, in the case of scintillation yields, blocking the source not only partially reverses the effect of the fast component, but causes a permanent yield reduction, the origin of which is unknown.

A further experiment on the radiation damage of CsI scintillator photocathodes measurements took place in May 1996, where both photoelectric and scintillation light yields were measured simultaneously. The driving force behind these measurements was to discover whether a CsI based scintillator combined with a CCD light detector is feasible as a detection system for synchrotron protein crystallography studies (Nagarkar et al. 1996).
Figure 8.13: The variation of scintillation light yield from thallium doped caesium iodide with accumulated dose at the two marked X-ray energies. The gap between measurements represents a period when the X-ray beam was not incident on the sample.

8.5 Discussion

While there are obvious discrepancies in the data, mainly the highly variant absolute efficiency values and some evidence of pre-irradiation of supposedly fresh sample regions, there are some strong general features in the measurements, the physical causes of which are not obvious. Effects such as higher energy photons causing less efficiency degradation, while the angle of incidence makes little difference can be understood by the qualitative descriptions above. A deeper understanding of the radiation damage process is required to appreciate the more complex causes of the dose rate effects and recovery characteristics. Chapter 9 investigates how changes at the atomic level caused by X-ray interactions might negatively affect the likelihood of photoemission. The remainder of this chapter describes an analysis of the variation, with angle of incidence and photon energy, of the constants in equation 8.1 giving the best fit to the measured decay scans.
8.5.1 Curve Fitting analysis

Various functions were fitted to the shape of the quantum efficiency versus accumulated dose curves, but an equation of the form of eq. 8.1 was found to be most accurate.

\[
\text{Photoyield}(F) = a_1 + a_2e^{-c_1 F} + a_3e^{-c_2 F}
\]  

(8.1)

where \( a_1 \) and \( c_i \) are constants and \( F \) is the accumulated flux.

Anderson et al. (1992) had also found a curve produced by summing two exponential components and a baseline was adequate to describe their measurements of CsI quantum efficiency variation with electron dose; while Levy (1991) required three or four exponential components to produce a best fit for \( f \) centre growth curves in alkali halides under X-ray irradiation.

A flow chart representing the procedure for optimising the fit between the measured quantum efficiency variation with time and equation 8.1 is shown in figure 8.14. A two component method was found to be most successfully in producing close fits, the first component accurately tying down the region in which the optimum should be found and the second finding the ideal values. Often, equally good fits could be found in very different regions of parameter space, but as can be seen in figures 8.15 and 8.16, the two stage procedure, more often than not, produces very similar fits for data sets where only the photon polarisation differed. Figure 8.15 examines the variation of fit parameters with angle of incidence for 2500 eV X-rays, and sheds some light on the angular variation seen in figure 8.9 where the samples were radiation harder at 25° than either 10° or 40°. The evidence from curve fitting analysis suggests that this radiation hardness is not due to an intrinsically slower time constant, just that both the decaying components are less prominent, a possible cause of which could be that the sample had previously been irradiated unintentionally. The general trend from 10° to 40° grazing incidence, ignoring the 25° data is then that the time constants increase, most probably as the flux per unit area does, but the prominence of the decay components decrease as more X-rays are absorbed deeper into the sample.

---

*An \( f \) centre is a type of lattice defect as shown in figure 9.1; its role in photodark degradation is discussed in the following chapter.*
Figure 8.14: Schematic of the procedure used to find the least squares fit between experimental decay scans and equation 8.1.
Figure 8.16 examines the variation of fit parameters with photon energy at 10° grazing incidence and a far stronger trend is immediately apparent whereby both decay component weightings and more especially time constants are reduced as the photon energy increases. Some caution must however be applied before too deeply analysing the possible causes of this, as the photon flux rate and beam spot size and shape (the 120eV data was obtained on a different beam line) are all energy dependent. It does, however, appear that the greater effect caused by deeper absorption depths is a reduced time constant, counter to the angular variation case above. My feeling was that this form of analysis is simply too basic to really understand the physical mechanisms behind the radiation damage processes. With that in mind an attempt to develop a detailed atomic model was made which is described in the following chapter.
Figure 8.15: The variation, with angle of incidence, of the parameters in equation 8.1 giving the optimum fit to the measured decay scans. +) baseline weighting \([a_1]\); *) weighting \([a_2]\) and time constant \([c_2]\) for the slower decay component; o) weighting \([a_3]\) and time constant \([c_3]\) for the fast decay component. Photon energy: 2500 eV
Figure 8.16: The variation of best fit parameters in equation 8.1 with photon energy at 10° grazing incidence. + ) baseline; *) first decay component; o) second decay component.
Chapter 9

Modelling of Radiation Damage

9.1 Introduction

Research in three fields appears crucial to our understanding of the flux-induced efficiency degradation in alkali halides. Fortunately, each of these—X-ray induced defect formation, annihilation and diffusion, surface desorption and photoemission mechanisms—have been extensively researched for alkali halides, as we shall see below. One of the main problems in relating this research to our measurements is that different alkali halides have aroused greater interest in different fields. CsI studies have generally concentrated on quantum efficiency for photoemission while virtually all the studies on colour centres and surface effects have been on crystals not containing caesium. Most alkali halides adopt the NaCl crystalline structure with a face centred cubic lattice structure with 6 nearest neighbour atoms, whereas caesium halides form a body centred cubic structure with 8 nearest neighbour atoms. The effect of this structural difference on defect mechanisms is unclear. The structure and mobility of colour centres in the two structures would be expected to differ, but the overall shape of quantum efficiency degradation curves are remarkably similar. Furthermore, a number of studies successfully extrapolate alkali halide centre energies to include caesium halide results (Itoh & Tanimura 1990). With the bulk of colour centre data relating to face centred cubic structured alkali halides I shall address the initial modelling effort to KBr; how it might then relate to CsI is addressed in section 9.7. The effects of impurities and photoemission modelling are assumed to apply equally to all alkali
halides.

9.2 X-ray Induced Colour Centre Formation

The effects on alkali halides of exposure to X-rays and electrons is a much studied topic. It has been known since 1930 that X-ray irradiation created colour centres (Mott & Gurney 1940), defects in the ionic crystal structure so named because their creation caused characteristic absorption bands in the visible region of the spectrum. The main absorption band formed at room temperature was denoted the F-band caused by absorbing F-centres (German Farbzentren or colour centres). The properties of these colour crystals have since been extensively studied, for example by Pohl (1937), Pooley (1966), Itoh & Tanimura (1990), Levy (1991) and Comins et al. (1994), leading to the elucidation of the nature of the F centre and of two connected entities important in the radiation damage process, the self trapped exciton and the H centre. Figure 9.1 illustrates the structure of the self trapped exciton and a pair of F and H centre defects. The F centre is the halogen ion vacancy that has captured an electron; the H centre is the result of a neutral halide atom arriving at an interstitial and covalently bonding with the halogen ion at that site. F centres can be created by trapping of an electron at an already present vacancy, but we are most interested in the creation of an F-H pair, the halogen atom creating an F centre as it is dislodged to an interstitial where it forms a H centre. It is currently accepted that X-ray absorption actually creates a self trapped exciton (figure 9.1a) which, via a displacement of the $X^-$ ion (X representing a halogen atom) without changing its electronic state, becomes an F-H pair. The self trapped exciton (STE) is truly an isomer of the F-H pair (Itoh & Tanimura 1990)

Itoh also found that the energy of the lowest STE state is slightly larger or very close to the energy of the F-H pair so it appears the transition from STE to F-H pair is exothermic.

At room temperature H centres are not observed because their thermal motion leads them to either recombine with F centres or form large interstitial complexes. In fact, motion of both H and F centres is found to lead to the creation of a large number of new absorption bands as defects self-trap. F centres may precipitate into $F_2$ (=M), $F_3$ (=R) and $F_4$ (=N) centres (Soppe & Prij 1994). With further growth these defect centres effectively become alkali metal "colloids" with the possibility of a significant region of the photocathode bulk becoming devoid of...
the halogen species. Trapping of H centres occurs mainly at impurities, with the formation of $S_1$, $S_2$ and $S_3$ centres consisting of an impurity with 1, 2 and 3 H centres respectively. Finally there are dislocation loops (figure 9.2), which are considered to be seeded by $S_3$ centres. According to Hobbs, Hughes & Pooley (1973) two H centres near a dislocation will form a halogen molecule by "digging out" say a KI molecule from its regular position. The KI molecule moves to the extra plane of the dislocation giving rise to growth of perfect dislocation loops as seen in figure 9.2 for both KI and KBr.

### 9.3 Surface Effects And Desorption

The formation within the photocathode bulk of a large number of mobile defects means that some of these will reach and interact with the sample surface. In characterising the likely effects of these interactions one can draw on the observations of alkali halide surfaces after irradiation and a number of studies of material desorption during irradiation.
Various measurements of desorption yields of neutral and ionised atoms from alkali halides under photon and electron stimulation have been conducted and although some results are contradictory a general picture as described below emerges. The emission of excited (Bunton et al. 1991) and ionised (Tokutaka et al. 1970) alkali atoms account for only a small fraction of the total desorption yield, hence their effect on the surface stoichiometry can be ignored. Many of the interesting desorption experiments have been conducted under bombardment by electrons of a few hundred eV; although the degree of damage caused by electrons is significantly greater than that caused by X-rays the processes of defect formation, diffusion and surface metallisation are believed to be sufficiently similar that these results are relevant to photon stimulated desorption, where far fewer experimental studies have been conducted.

Loubriel et al. (1986) and Green et al. (1987) studied the time variation of Li desorption from LiF under pulsed electron bombardment and found F centre diffusion to be the rate limiting factor for Li desorption rather than thermal evaporation. Considering the nature of the F centre it indeed seems likely that surface interaction would result in the neutralisation of a surface alkali atom, which could be immediately ejected from the sample or remain on it, perhaps thermally desorbing later. At low temperatures an alkali metal overlayer has the potential to form which
would tend to inhibit further desorption (Sarnthein et al. 1991) and secondary electron emission. Such overlayers have been witnessed on a variety of alkali halide species (Lord & Gallon 1973; Roy, Singh & Gallon 1985). Most importantly in connection with this study, Dangendorf et al. (1991) found that on exposure to air of a CsI photocathode the irradiated spots immediately turned black as the Cs overlayer was oxidised. A similar consideration of the nature of the H centre suggests that on meeting the surface the extra neutral halogen that makes up the H centre will be released from the sample. However, since this atom would have been carried from deep within the photocathode, the surface stoichiometry is unlikely to be affected. Tokutaka et al. (1970) measured yields of neutral halogen species to be between 5 and 10 times greater than the alkali metal yields as would be expected if their emission resulted from the surface interaction of highly mobile H centres. Similarly, the measurement of desorption versus photon energy made by Szymondski et al. (1992) from NaCl in the region of the Cl-K absorption edge (2.833keV) can be explained by this model. Neutral sodium desorption ¹ was found to exactly follow photoemission across the absorption edge showing that the depth at which F centres are created is as important to desorption yield as photoelectron creation depth is to photoemission.

Finally, figure 9.3 shows the variation with temperature of lithium desorption yields under X-ray and electron bombardment. Sarnthein et al. (1991) using electron bombardment, split the yield behaviour into four temperature ranges, below 150°C the vapour pressure of the alkali metal was negligible and no desorption occurred, above 150°C the yield increased exponentially until 300°C where the desorption rate equalled the lithium metal formation rate. Measured desorption only increases again above 450°C when evaporation of the LiF crystal begins. Husinsky et al. (1988) using X-ray illumination found a similarly shaped increase in ground state ² yield but at a completely different temperature (300°C c.f. 150°C). With both experiments conducted under high vacuum conditions this anomaly is hard to explain. The shapes of both yield versus temperature curves are however consistent with the hypothesis of the formation of an alkali metal overlayer. Similar data is unavailable for Cs or K overlayers but these heavier alkali metal atoms at room temperature maintain vapour pressures equal to that of lithium at 300°C for caesium and 230°C for potassium. Not knowing which, if either, of the lithium experiments is applicable to our measurements, we can only estimate levels of likely Cs desorption from the

¹Although halogen yields are generally much higher than yields of the metal they are far harder to measure and thus many studies including this one only record metal yields.

²Ground state refers to the desorption of low energy neutral atoms from the surface.
Figure 9.3: The variation in yields of desorbed Li from LiF with temperature. Left: electron stimulated desorption data of (Sarnthein et al. 1991). Right: photon stimulation desorption data of (Husinsky et al. 1988), \( \text{Li}^\circ \) represents the ground state neutral Li yield and \( \text{Li}^* \) the yield of excited lithium atoms.

9.4 Photoemission From A Degraded Sample

None of the previously introduced defects or changes in structure or stoichiometry are directly measured by the experiments described in the previous chapter. The photoelectric yields will however depend on the effects and concentration of many of them. The formulation for photoyields used here follows that of Fraser (1983a) where the secondary electron contribution to the current quantum yield is given by:

\[ Y = Y_0 	imes \left( \frac{1}{1 + \frac{I}{I_0}} \right) \]

where \( Y \) is the photoyield, \( Y_0 \) is the primary electron yield, \( I \) is the primary electron current, and \( I_0 \) is a reference current.
\[ (\chi_c)_s = [(1 - R(\alpha)).f.P_s(0).E_x.\epsilon^{-1}(1 + \beta)^{-1}]\left[1 - e^{-\left(\mu \text{cosec} \alpha'.L_s\right)}\right] \]  

(9.1)

where \( R(\alpha) \) is the reflectivity at an angle of incidence \( \alpha \)
- \( f \) the conversion efficiency of photon energy to energy of secondary electrons
- \( P_s(0) \) the probability of the \( e^- \) escaping into vacuum, having reached the surface
- \( E_x \) the photon energy
- \( \epsilon \) the energy required to free a secondary electron
- \( L_s \) the secondary electron escape length
- \( T \) is the thickness of the alkali halide layer.
- \( \beta = (\mu \text{cosec} \alpha'.L_s)^{-1} \) where,
- \( \mu \) is the linear absorption coefficient.
- and \( \alpha' \) the angle after surface refraction

The second term in the above formula accounts for the fact that the alkali halide layer may not be thick enough to absorb all the incident X-ray flux.

The total current quantum yield, that is the total number of electrons released from the sample per incident photon, is \((\chi_c)_p\) plus \((\chi_c)_p\), the primary photoyield. An analytical form for \((\chi_c)_p\) is also given by Fraser (1983a), but because for CsI only a few percent of the total photoyield consists of primary electrons (Henke, Knauer & Premaratne 1981) and the primary photoyield would be less affected by the formation of defects or an alkali metal overlayer, \((\chi_c)_p\) will be treated as a small constant i.e.

\[ (\chi_c) = 0.05(\chi_c)_{\text{CsI}} + [(1 - R(\alpha)).f.P_s(0).E_x.\epsilon^{-1}.(1 + \beta)^{-1}]\left[1 - e^{-\left(\mu \text{cosec} \alpha'.L_s\right)}\right] \]  

(9.2)

where \((\chi_c)_{\text{CsI}}\) is the photoyield from pure, undamaged CsI.

Examinations of the quantum efficiency decay in alkali halides, such as those by Lu et al. (1995) and Breskin (1996) have claimed that surface changes cause the rapid decay component dominating the decay process. If this is so, the most significant term in equation 9.1 is \( P_s(0) \), the probability of an electron having reached the surface, penetrating it. In CsI a value for \( P_s(0) \) of 0.2 gave good fits to a number of experimental results (Fraser 1983b) while from the data of Ono

---

3 Primary and secondary photoyields are distinguished by the electron energies as illustrated by figure 2.8
& Kanaya (1979) 0.023 would be more reasonable for Cs. Thus, the formation of a complete Cs overlayer has the potential to reduce quantum efficiency measurements by almost an order of magnitude. Single defects formed in the bulk have been shown to significantly alter the electron escape depth, \( L_e \). Pohl (1937) found that F centres efficiently trapped photoelectrons which, as illustrated in figure 9.4 for KCl, led to a linear reduction in the range of electrons as the F centre concentration increased. From this data Pohl calculated the cross section of a single F centre in KCl to be \( 4 \times 10^{-17} \text{m}^2 \) with cross sections of the same order of magnitude measured for KBr, NaCl, RbCl and RbBr.

The significant differences in experimental parameters between Pohl’s experiment and our own means it is unlikely that this value for the cross-section is directly transferable to our model. Pohl produced a stoichiometric excess of alkali metal (mainly as F centres) by heating crystals in the presence of the cation metal and the electrons, whose ranges were measured, were produced by irradiation of the crystal with optical light in the F band and subjected to an electric field to drag them toward an electrode. This differs in three important ways from the X-ray radiation damage measurement of Chapter 8. Most importantly, the number of electrons is always much less than the number of F centres which were their source; secondly, some electrons will be trapped not in F centres but by halogen vacancies [a halogen vacancy being an F centre without

Figure 9.4: The effect of F-centres on the electron escape escape depth in KCl (Pohl 1937)
a trapped electron and thus positively charged]. Thirdly, Pohl's measurements were conducted at -100°C.

X-ray irradiation of KBr or CaI produces more electrons than F centres so saturation would likely occur on reasonably short timescales destroying the linear relationship. The second point is probably not significant, as the relationship between F centre concentration and electron range is linear over a wide range of F centre concentrations and although an F centre is nominally neutral, the wave function of the single electron trapped at the F centre extends over a greater volume than that of the negative ion it replaces. Thus there will be a small potential hole in this volume capable of trapping a second electron (Mott & Gurney 1940). The third point is significant for at higher temperature the electrons are less likely to be trapped at these only very slightly positively charged sites. Mott & Gurney (1940) give a temperature relationship for the cross section as;

$$\sigma = \left(\frac{\omega}{F}\right)^{-1} e \frac{I}{\delta kT N}$$

(9.3)

where $\omega/F$ is the electron range per unit electric field, which increases 100 fold between -100°C and room temperature (Pohl 1937); $I$ is the mean free path which falls as $e^{\Omega/kT}$ as $T$ rises ($\Omega = h\nu/k$ taken as 800K) and $N$ is the F centre concentration. Correcting for temperature in this manner the F centre cross section, $\sigma_F$ is reduced to $3.10^{-14}$ m$^2$. While the issue of F centre saturation must be considered in interpreting the model's results, a value for the F centre cross section of $3.10^{-14}$m$^2$ was adopted as an upper limit in the absence of more applicable data. From this value of the cross section, an electron mean free path due to F centres, $\lambda_F$ was calculated by;

$$\lambda_F = \frac{I}{\sigma_F} = \frac{M_o}{\rho N_A \sigma_F \epsilon_F}$$

(9.4)

where $\rho$ is the material density, $M_o$ its atomic mass, $N_A$ Avogadro's number and $\epsilon_F$ the F centre mole fraction. The true electron escape length, $L_e$ in equation 9.1, is then found from,

$$\frac{1}{L_e} = \frac{1}{L'_e} + \frac{1}{\lambda_f}$$

(9.5)

where $L'_e$ is the escape escape length for the fresh alkali halide. For CaI this is taken as 250Å after Henke (1980) and as 160Å for KBr by comparison with other potassium halides examined by Fraser et al. (1983).
Alkali colloids and dislocation loops could also affect the linear absorption coefficient ($\mu$), energy required to produce secondary electrons ($\epsilon$) and the electron escape depths. A basic interpretation of the effect of alkali colloids is to assume they behave as the pure alkali metal would in that small volume region. The photocathode bulk is thus treated as if it were part CsI with an increasing proportion of Cs and the bulk parameters varying as the colloid (Cs) volume increases; i.e.

$$\epsilon(t) = (1 - f_B)\epsilon_{Cs} + f_B\epsilon_{Cs}$$

and

$$L_s(t) = (1 - f_B)L_s[CsI] + f_BL_s[Cs]$$

where $f_B$ is the fractional volume taken up by colloids. Under all parameter sets analysed here $f_B$ remains so small ($< 10^{-30}$) that the effects of colloids on these parameters is negligible.

### 9.5 The Jain - Lidiard - Soppe Model

No systematic study of the build up of colour centres in CsI nor KBr has ever been conducted. Levy (1991) has examined the variation of $F$ centre concentrations during and after photon irradiation for KCl and Comins et al. (1994) produced an empirical model for the variation in $F$-centre concentration in the same material. Both showed $F$-centre growth followed the form;

$$f(t) = \sum_{j=1}^{2} A_j(1 - e^{-\lambda_j t})$$

where $t$ represents time and $A_j$ and $\lambda_j$ are positive real numbers based on the various model parameters or found by best fit to the experimental data. The model of Comins et al. (1994) is however not particularly useful when considering the surface effects as colour centre diffusion is avoided. Far more attention has, however, been paid to defects in NaCl as deep rock salt formations have been proposed as possible permanent repositories for high-level radioactive waste. One of the major concerns for the safety of such repositories is that of an explosive back-reaction between colloids and molecular chlorine induced by photon irradiation. To see if the concentrations of these defects would ever reach such back reaction thresholds, Jain & Lidiard (1977) developed a quantitative model of colour centre and colloid formation, diffusion and
annihilation, which has since expanded by Soppe (1993) to cover all the defect types described at the beginning of this chapter and the various possible reactions between them as marked by the dashed lines in figure 9.5.

The rate equations given by Soppe (1993) for F centre concentration in NaCl is, 

\[
\begin{align*}
\frac{dc_f}{dt} &= K - k_{2F}c_{H} + 2k_{M}c_{M} - 2k_{F}c_{H}c_{F} - k_{F}c_{F}c_{M} - k_{F}c_{F}c_{R} - k_{F}c_{F}c_{N} \\
&+ k_{H}c_{H}c_{M} - 4\pi r_{c}C_{D}(c_{F} - c_{F}^{eq}) - 2\rho_{F}D_{F}c_{F} - \gamma D_{F}c_{F}c_{z_{1}}
\end{align*}
\]  

(9.9)

with parameters as defined in table 9.1. The first term, K, is the production rate of F-H pairs from self trapped excitons, the second term annihilation between F and H centres, the third term accounts for the disintegration of an M centre into two F centres. The next four terms describe the trapping of F centres at F, M, R and N centres respectively. The eighth term accounts for F centres formed from the back reaction between H and M centres. The term \(4\pi r_{c}C_{D}(c_{F} - c_{F}^{eq})\) refers to the loss of F centres to colloids and the evaporation of F centres from colloids, and the last two terms represent F centres trapped at dislocation loops and by molecular chlorine respectively.

Similar differential equations are valid for the remainder of defect species,

\[
\begin{align*}
\frac{dc_{H}}{dt} &= K - k_{2F}c_{F} - k_{H}c_{H}c_{S_{0}} - k_{H}c_{H}c_{S_{1}} - k_{H}c_{H}c_{M} - k_{H}c_{H}c_{S_{1}}c_{H}c_{S_{1}}
\end{align*}
\]
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$c_F$</td>
<td>mole fraction of F centres</td>
</tr>
<tr>
<td>$c_H$</td>
<td>fraction of H centres</td>
</tr>
<tr>
<td>$c_M$</td>
<td>fraction of M centres</td>
</tr>
<tr>
<td>$c_R$</td>
<td>fraction of R centres</td>
</tr>
<tr>
<td>$c_N$</td>
<td>fraction of N centres</td>
</tr>
<tr>
<td>$c_A$</td>
<td>fraction of Na in colloids</td>
</tr>
<tr>
<td>$c_S_0$</td>
<td>fraction of empty H centre traps</td>
</tr>
<tr>
<td>$c_S_1$</td>
<td>fraction of single H centres trapped at impurities</td>
</tr>
<tr>
<td>$c_S_2$</td>
<td>fraction of H dimers trapped at impurities</td>
</tr>
<tr>
<td>$c_S_3$</td>
<td>fraction of H trimers trapped at impurities</td>
</tr>
<tr>
<td>$c_Cl_2$</td>
<td>fraction H centres converted to molecular Cl$_2$</td>
</tr>
<tr>
<td>$c_F$</td>
<td>fraction F centres in equilibrium with colloids</td>
</tr>
<tr>
<td>$c_H$.S$_1$</td>
<td>fraction H centres in equilibrium with S$_1$ centres</td>
</tr>
<tr>
<td>$c_H$.S$_2$</td>
<td>fraction H centres in equilibrium with S$_2$ centres</td>
</tr>
<tr>
<td>$K$</td>
<td>production rate of F-H pairs</td>
</tr>
<tr>
<td>$k_2$</td>
<td>rate constant for recombination of F and H centres</td>
</tr>
<tr>
<td>$k_F$</td>
<td>rate constant for formation of M centres and colloids</td>
</tr>
<tr>
<td>$k_R$</td>
<td>rate constant for formation of R and N centres</td>
</tr>
<tr>
<td>$k_H$</td>
<td>rate constant for formation of S$_n$ clusters</td>
</tr>
<tr>
<td>$K_M$</td>
<td>dissociation rate of M centres</td>
</tr>
<tr>
<td>$D_F$</td>
<td>diffusion coefficient for F centres</td>
</tr>
<tr>
<td>$D_H$</td>
<td>diffusion coefficient for H centres</td>
</tr>
<tr>
<td>$r_c$</td>
<td>mean radius of Na colloids = $\sqrt{(3c_A/4\pi C_c)}$</td>
</tr>
<tr>
<td>$C_c$</td>
<td>number of colloids per cm$^3$</td>
</tr>
<tr>
<td>$s_H$</td>
<td>dislocation bias coefficient for H centres</td>
</tr>
<tr>
<td>$s_F$</td>
<td>dislocation bias coefficient for F centres</td>
</tr>
<tr>
<td>$\rho_d$</td>
<td>dislocation line density</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>rate constant for back reaction between F centres and molecular Cl$_2$</td>
</tr>
<tr>
<td>$N$</td>
<td>number of Na$^+$ ions per cm$^3$</td>
</tr>
<tr>
<td>$C_l$</td>
<td>number of dislocation loops per cm$^3$</td>
</tr>
<tr>
<td>$C_l^0$</td>
<td>number of nuclei for dislocation loops per cm$^3$ at time t=0</td>
</tr>
<tr>
<td>$b$</td>
<td>Burgers' vector of dislocation loops</td>
</tr>
<tr>
<td>$\nu$</td>
<td>ratio of molecular volume of NaCl and atomic volume of metallic Na</td>
</tr>
</tbody>
</table>

Table 9.1: Symbols used in the Jain - Lidiard model
The rate equation for the concentration of empty II centre traps is:

\[
\frac{dc_M}{dt} = k_{PCP} - k_{HCHCM} - k'_{CM} - k_{HCPM} \tag{9.10}
\]

\[
\frac{dc_R}{dt} = k_{HCFR} - k_{HFCR} \tag{9.11}
\]

\[
\frac{dc_N}{dt} = k_{HCFR} - k_{HFCN} \tag{9.12}
\]

\[
\frac{dc_A}{dt} = k_{HCFN} + 4\pi r_c A (D_{PCP} - D_{HCH} - D_{PFC}) \tag{9.13}
\]

\[
\frac{dc_{GA}}{dt} = k_{HCEH} - k_{HCEH} - k_{HCEH} \tag{9.14}
\]

\[
\frac{dc_{GA}}{dt} = k_{HCEH} - k_{HCEH} - k_{HCEH} \tag{9.15}
\]

\[
\frac{dc_{GA}}{dt} = k_{HCEH} - k_{HCEH} \tag{9.16}
\]

\[
\frac{dc_{GA}}{dt} = k_{HCEH} - k_{HCEH} \tag{9.17}
\]

\[
\frac{dc_{GA}}{dt} = k_{HCEH} \tag{9.18}
\]

\[
\frac{dc_{FA}}{dt} = \rho_f (z_H D_{HCH} - z_P D_{PCP}) - \gamma D_{PFC} \tag{9.19}
\]

The rate equation for the concentration of empty II centre traps is:

\[
\frac{dc_{SA}}{dt} = k_{HCEH} c_{HCH} - K_{HCEH} c_{HCH} \tag{9.20}
\]

which has the initial value (at t=0) of \( c_{SA} \), the concentration of impurities. A final differential equation is required to track the colloid concentration as an N centre trapping another F centre is considered to have become a new colloid. Hence;

\[
\frac{dC_l}{dt} = N k_{PCP} \tag{9.21}
\]

New dislocation loops can also be formed by the mechanism described by Hobbs, Hughes & Pooley (1973). An \( S_3 \) centre is considered a dislocation loop seed so that;

\[
C_l = C_l^0 + N c_{SA} \tag{9.22}
\]

Finally, we use Jain and Lidiard's relation between the dislocation line density and the number of molecular chlorine centres;

\[
\rho_d = \sqrt{(2\pi \rho_0/b)} \tag{9.23}
\]

where \( \rho_0 \) is the dislocation line density at time t=0. The basic model, which produced reasonable fits to measurements of colloid growth and molecular chlorine centres in NaCl (Soppe
was then extended, as described below, to deal with the additional effects relevant to our photoemission experiments. The resulting system of rate equations was solved using the Isoda package developed at the Lawrence Livermore Laboratory. The solver is particularly useful as it determines internally if the system of ordinary differential equations (ODEs) is stiff or non-stiff and chooses the appropriate method to solve them (Petzold 1983). Over the range of input parameters used the system turned out to be stiff in all cases.

9.6 Extensions to the Jain - Lidiard - Soppe Model

The model presented above takes no account of the processes that occur prior to the separation of F-H centre pairs, instead simply assuming a steady rate, K, are produced. Although this is adequate for any of our single synchrotron measurements lasting a couple of hours, K will be affected by both changes in the photon energy and angle of incidence. The next two sections deal with this problem and even more importantly how the model is extended to deal with the interaction between mobile defects and the sample surface.

9.6.1 The Variation of F and H Centres Production Rates With Angle of Incidence and Photon Energy

The definition of K, the production rate of F and H centres, is per atom; i.e.

\[ K = \frac{\text{no. absorbed photons}}{\text{no. of atoms}} \cdot \frac{\text{photon energy}}{\text{F - H pair production energy}} \]  

(9.24)

Figure 9.6 illustrates the significant regions of the photocathode during grazing incidence measurements. The dark shaded area represents the main region for both F-H pair and photoelectron creation. F centres and electrons are also released in the lighted shaded region but this is considered too deep within the sample for either to reach the surface. This depth, denoted z in figure

4 A stiff system is one where terms within one equation have very different timescales leading to instabilities which restrict a numerical method to very small time steps. In mathematician jargon: Over an interval [a,b] an ODE of the form \( y' = f(x,y) \) is said to be stiff if in the neighbourhood of a solution \( y \) there exists a component of \( y \) whose variation is large compared to \( (b-a)^{-1} \).
Figure 9.6: Regions of interest within the irradiated photocathode. See accompanying text for details. The dark shaded area represents the main region for both F-H pair and photoelectron creation. F centres and electrons are also released in the lighted shaded region but this is considered too deep within the sample for either to reach the surface.

9.6, is considered constant for all experimental parameters. Defect diffusion into the unshaded region will mean that there is a reasonable F centre concentration here too, but the model, unable to deal with physical diffusion in this way, is forced to assume defect concentration is constant within this surface layer. As this is obviously inaccurate most model predictions will be conducted at normal incidence where the assumption is more reasonable. With $z=2000\,\text{Å}$ and a beam size of 4mm by 1mm (the spot size of SRS beamline 6.3), the significant volume in which defect production occurs is $4a\,\text{mm}^3$ where $a=z[\text{mm}]/\sin\theta$. The number of atoms of KBr in this volume is then $2.21 \times 10^{19}/\sin\theta$ and the number of CsI atoms $1.73 \times 10^{19}/\sin\theta$. The energy required to create a F-H pair in KBr is taken as 100eV by extrapolating the low temperature measurements of Sonder (1975) to room temperature and rather arbitrarily in the absence of measurements to be 80 eV for CsI. The numbers of photons absorbed were calculated from the linear absorption coefficients, $\mu$, where,

$$\mu = \frac{4\pi\beta}{\lambda}$$

(9.25)

where the optical constant, $\beta$ (actually the complex component of the refractive index) is itself calculated from the atomic scattering factor $f_g$.

$$\beta = \left( \frac{r_0\lambda^2}{2\pi} \frac{N_A}{A} \rho \right) f_g$$

(9.26)

$f_g$ is obtained by applying equation 9.27 to selected parameters within the photoelectric cross-section database of Cromer & Liberman (1970).
Table 9.2: F-H pairs creation per in beam atom per X-ray in CsI and KBr at various grazing angles of incidence and photon energies. Blanks imply no measurements corresponding to these parameters were conducted.

\[
f_2(E) = \frac{E\mu(E)}{2\pi r_0 \hbar c}
\]

(9.27)

where \( r_0 \) \( =e^2/4\pi\epsilon_0 m_e c^2 \) = 2.818x10^{-15}m \) is the classical electron radius, \( N_A \) Avogadro’s number and \( A \) and \( \rho \) are the atomic weight and material density respectively.

Based on equation 9.24 the values of \( K \) found by using the above calculations are displayed in table 9.2. The actual values entered into the computer model are found by multiplying these \( K \) values by a time averaged beam strength in photons per second and are displayed in table 9.3.

### 9.6.2 Defect - Surface Interaction

Technically, defect diffusion from the X-ray interaction region to the surface should be modelled by the inclusion of second order differential terms in the equations 9.9 and 9.10. Knowing no method of solving such a system of equations, simpler surface terms were included effectively treating the surface as a very large number of dislocation lines, at least as far as the diffusion of defects onto it were concerned. A new parameter, \( S_m \), the surface metallisation, was introduced to the model, with the corresponding rate equation,

\[
\frac{dS_m}{dt} = \left(1 - S_m(1 - u) - uS_m^2\right)D_{FVFP} - gS_m
\]

(9.28)
Table 9.3: F - H pairs creation per in beam atom per second for CsI and KBr at various grazing angles of incidence and photon energies assuming an average beam strength from measurements with these parameters. Blanks imply no measurements corresponding to these parameters were conducted.

<table>
<thead>
<tr>
<th>Daresbury Beamline</th>
<th>Material</th>
<th>Angle</th>
<th>Photon Energy (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>2000</td>
</tr>
<tr>
<td>6.3</td>
<td>CsI</td>
<td>10°</td>
<td>1.58.10^-6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>25°</td>
<td>1.89.10^-6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>40°</td>
<td>2.00.10^-6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>90°</td>
<td>2.14.10^-6</td>
</tr>
<tr>
<td>3.4</td>
<td>CsI</td>
<td>90°</td>
<td>1.15.10^-6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>90°</td>
<td>5.40.10^-7</td>
</tr>
<tr>
<td></td>
<td>KBr</td>
<td>90°</td>
<td>1.30.10^-7</td>
</tr>
</tbody>
</table>

It is assumed that when an F centre reaches the surface it will neutralise an alkali ion, unless it meets a surface region already covered by a metal overlayer in which case the process of the F centre diffusing through the sample will continue with a possibility, u, that it will again meet a surface region, which may or may not be metalised. For the model results presented below u is given an arbitrary value of 40%. The second term accounts for evaporation of metal atoms; where \( g \) is the probability of each surface atom evaporating in any given second. Equations 9.9 and 9.10 were also adapted so as to account for the loss of F and H centres at the surface:

\[
\frac{dF}{dt} = \left( \frac{dF}{dt} \right)_{dis} - (1 - S_m)D_F c_F \rho_s
\]

(9.29)

and

\[
\frac{dH}{dt} = \left( \frac{dH}{dt} \right)_{dis} - D_H c_H \rho_s
\]

(9.30)

In all cases \( \rho_s \) is a dislocation line density type term representing the surface rather than internal dislocations.
9.7 Modelling Radiation Damage in CsI

Raman spectroscopy measurements have shown the association of H centres to form $X_3^-$ and larger aggregates occurs equally well in Cs compounds as sodium or potassium ones, but the stabilisation of halogen molecules into aggregates of the $(X_2)_n$ form occurs only in crystals where the cation has a mass equal or below that of potassium (Bernard, Rzepka & Lefrant 1990). Overall, this is good news and means the Jain-Lidiard-Soppe model can be reasonably applied to CsI as other alkali halides, with parameter values so as to make halogen molecule aggregation unlikely.

9.8 The Role Of Thallium Dopants In Alkali Halides

The photoyield measurement of figure 8.12 shows that thallium doped caesium iodide is less radiation hard than pure CsI, while Al Janmal, Pooley & Townsend (1973) found Tl doping reduced the yields from electron induced desorption from KI. This section addresses the question as to whether, in terms of the model described above, a single effect can explain both apparently contradictory results. Al Janmal, Pooley & Townsend (1973) ascribed the increased stability of Tl doped samples to the view that they acted as recombination centres for diffusing excitons, thus reducing the number of such defects that reach the surface and cause sputtering. Al Janmal claims F centre diffusion would not be affected by Thallium ions and thus cannot be the cause of the reduction in sputtering efficiency. This explanation though cannot explain the greater photoyield degradation from doped samples. One possibility is that Tl acts as an anchor for the formation of metal islands on the sample surface. This would both reduce sputtering and quantum efficiency. Limited support for this proposal comes from a measurement by Vollmer & Träger (1987) of cluster sizes and distributions of Na atoms deposited on LiF. They found a wide range of metal island sizes, but a density of $3.10^6\text{cm}^{-2}$, a number typical of the layer defect density for alkali halide crystals of the type used. Another possibility is thallium acts like the impurities described by Soppe (1993) in the above defect model, i.e. they trap the highly mobile H centres preventing their recombination with F centres (Agullo-López, López & Jaque 1982). Sonder & Sibley (1965) indeed saw an enhancement in F centre production resulting from extra impurities; an effect which could cause enhanced quantum efficiency degradation due to a
reduction of electron range as described in section 9.4. Increased H centre trapping would also reduce halogen desorption although it is difficult to say whether the increased alkali desorption would allow this model to tie in with the results of Al Jammal, Pooley & Townsend (1973), but possibly because this desorption was studied under electron bombardment a different mechanism exists anyhow.

9.9 Comparison with Photoconversion Measurements

The Jain-Lidiard model tracking the concentration of various defects with accumulated flux produces results of the type displayed in figure 9.7. This figure assumes that the photocathode has the absorption and STE production properties of KBr, the defect diffusion and colloid formation characteristics of NaCl as assumed by Soppe (1993) (values for KBr are not known) and the X-ray flux of SRS beamline 3.4 through the monochromator set to 2000eV. These parameters are as set out in the first column of table 9.4.

The concentration of F centres can be seen to rise linearly until it reaches a plateau concentration of 500 parts per million when annihilation, formation of M centres and surface interaction balance the F centre production rate. The growth of F centres predicted by the model is similar in shape to the measurements of Levy (1991), although Levy found the F centre concentration in crystals continued to grow slowly after the period of rapid growth ended. This could be the result of an increase in the dislocation line density with accumulated flux, which trap more H centres reducing F-H annihilation. Such an effect is less significant in thin photocathodes as defect interaction with the surface swamps that with dislocation lines. The model predicts a H centre concentration remaining approximately constant with time. The highly mobile H centres are quickly annihilated upon meeting F accumulates or trapped by other imperfections never allowing a build up of this species. In fact, as time passes and the concentrations of other defects increase, H centres are annihilated or trapped even more rapidly leading to further reductions in their concentration. M centre concentration obeys the well known relationship $c_M \propto c_F^3$ (Schulman & Compton 1962). The main other defect species are impurities trapping 1 or 3 H centres (S1 and S3 centres). With the "NaCl like KBr" parameters the absolute values of defect concentrations predicted by the model are very interesting. After two hours irradiation
Figure 9.7: The time variation in concentration of the most significant defects and surface metalli­sation in KBr under a constant X-ray flux. Parameters in the Jain - Lidiard model are as given in the "NaCl like KBr" column in table 9.4. Defect diffusion and rate constants are as Soppe (1993) used for NaCl, but other parameters have the correct values for KBr. Both figures display the same data, the top graph using log-log axes and the lower graph linear axes.
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>Value used during model for NaCl like KBr</th>
<th>surface weighted KBr</th>
<th>Caesium Iodide</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D_F$</td>
<td>cm$^{-2}$s$^{-1}$</td>
<td>0.0015 $e^{-0.80/kT}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$D_H$</td>
<td>cm$^{-2}$s$^{-1}$</td>
<td>0.001 $e^{-0.10/kT}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$K_F$</td>
<td>s$^{-1}$</td>
<td>$10^{16} D_F$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$K_2$</td>
<td>s$^{-1}$</td>
<td>$10^{14} e^{-0.10/kT}$</td>
<td>2.10$^{13} e^{-0.10/kT}$</td>
<td>[1]</td>
</tr>
<tr>
<td>$K_R$</td>
<td>s$^{-1}$</td>
<td>$K_F e^{-0.20/kT}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$K_H$</td>
<td>s$^{-1}$</td>
<td>$10^{16} D_H$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$K_{eq}^M$</td>
<td>s$^{-1}$</td>
<td>$2.10^{-3} K_F$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$c_F^{eq}$</td>
<td></td>
<td>$2.10^{-4} e^{-0.30/kT}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$c_{H,S_1}^F$</td>
<td></td>
<td>$5.10^{-10} e^{-0.25/kT}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$c_{H,S_2}^F$</td>
<td></td>
<td>$5.10^{-10} e^{-0.10/kT}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$z_F$</td>
<td></td>
<td>1.0</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$z_H$</td>
<td></td>
<td>1.1</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>cm$^{-2}$</td>
<td>$10^{18} e^{-0.55/kT}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$C^0_c$</td>
<td>cm$^{-3}$</td>
<td>$10^{15}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$C^0_i$</td>
<td>cm$^{-3}$</td>
<td>$10^{15}$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$\rho_0$</td>
<td>cm$^{-2}$</td>
<td>$10^5$</td>
<td>[1]</td>
<td>[1]</td>
</tr>
<tr>
<td>$\rho_S$</td>
<td>cm$^{-2}$</td>
<td>$10^{12}$</td>
<td>$10^{12}$</td>
<td>[1]</td>
</tr>
<tr>
<td>$g$</td>
<td>s$^{-1}$</td>
<td>$2.10^{13} e^{-1.0/kT}$</td>
<td>$10^{12} e^{-1.0/kT}$</td>
<td>[1]</td>
</tr>
<tr>
<td>$N$</td>
<td>cm$^{-3}$</td>
<td>$1.39 \times 10^{22}$</td>
<td>[1]</td>
<td>1.05 $\times 10^{22}$</td>
</tr>
<tr>
<td>$b$</td>
<td>cm</td>
<td>$4.70 \times 10^{-8}$</td>
<td>[1]</td>
<td>5.65 $\times 10^{-8}$</td>
</tr>
<tr>
<td>$\nu$</td>
<td></td>
<td>0.96</td>
<td>[1]</td>
<td>0.82</td>
</tr>
<tr>
<td>$K$</td>
<td>s$^{-1}$</td>
<td>$5.40 \times 10^{-7}$</td>
<td>$5.40 \times 10^{-7}$</td>
<td>varied</td>
</tr>
</tbody>
</table>

As used in figure: 9.7 & 9.8

Table 9.4: Parameter values used in the various runnings of the Jain-Lidiard model. "NaCl like KBr" refers to the diffusion and rate constant values for NaCl as used by Soppe (1993), but known physical parameters relating to KBr. "Surface weighted KBr" fixes the relevant unknown parameters such as surface interaction dominates F centre interactions. The caesium iodide column is "NaCl like Csl". [1] implies the original value is used i.e. for NaCl like KBr.

173
at synchrotron fluxes the predicted surface coverage is less than 0.1% having virtually no effect on the efficiency of photoemission. On the other hand an F centre concentration of 0.05%, when inserted into equation 9.4, has a ridiculous large effect, reducing the electron mean free path to 0.2 Å. There are two main reasons for this; firstly the temperature dependence of the electron cross section is not as simple as suggested by equation 9.3 which is only truly valid in a limited temperature range, above which electrons achieve a higher thermally enhanced range as illustrated by figures 17 and 18 in Pohl (1937). For KBr and CsI, room temperature (20°C) is above this critical temperature and so the F centre cross section should be significantly below the value of $3 \times 10^{-14}$ m$^2$ originally assumed. Ideally measurements similar to Pohl’s would be conducted for KBr and CsI crystals but in their absence one can only estimate from the behaviour in other alkali halides the likely cross section reduction factor. Below I shall assume the cross section is an order of magnitude lower than that predicted by equation 9.3. This has been chosen as much to give reasonable values for the electron escape ranges as for how physically realistic a value it represents. Secondly the original assumption that a single F centre can trap an infinite number of electrons is absurd. In reality each F centre should only have the capacity to trap one electron and with far more electrons in the system than F centres, vacant F centres would be comparatively rare. Unfortunately the current model cannot combine electron and defect transport properties due to the way the series of differential equations is solved; to know the positions of electrons and defects at all times would require the addition of diffusion based second order differential terms within equations 9.9 to 9.21 making the system unsolvable. I shall therefore simply assume a constant fraction, 1%, of F centres are vacant (i.e. are as illustrated in figure 9.1, not having trapped a second electron). The combined effects of including these assumptions is that the electron escape length, $L_e$, is reduced from 160 Å for pure KBr to 95 Å as the F centre concentration rises to 500 parts per million, corresponding to a 40% reduction in quantum efficiency. Of course the model was based on parameters for defect interaction and diffusion in NaCl, an almost totally arbitrary selection of $\rho_a$ and an assumption that F centre concentration is equal in the irradiated and unirradiated regions.

Figure 9.8 examines the processes of F centre creation, annihilation, trapping by various defect species and surface interaction. The dominant process removing F centres from the sample is clearly F - H centre annihilation for these parameters. This is highly encouraging as it could explain some of our strangest experimental results especially why increasing the flux per...
Figure 9.8: The rates of formation and destruction of F centres in NaCl like KBr under a synchrotron flux characteristic of SRS beamline 3.4. The filled squares represent the production of F centres from X-ray absorption. The four mechanisms by which F centres are lost are vertically stacked to show after approximately 4000s the F centre concentration reaches equilibrium.

unit area had little effect on the rate of quantum efficiency degradation (i.e. figures 8.10 and 8.11). Under the model’s assumptions figure 9.8 would suggest the most likely effect of an increase in F centre production would be an even greater degree of F - H annihilation. A fuller examination of how the quantum efficiency degradations observed might be explained in terms of this model is conducted below. First, it is important to check that this conclusion isn’t highly dependent on the choice of input parameters and that by carefully selecting these the model cannot alternatively show surface effects dominate. Detector physicist have long held the view that alkali halide degradation is indeed surface effect dominant (Breskin (1996) and references therein.)

The dominance of annihilation over surface interaction leaves some scope for obtaining a higher surface metallisation by increasing $\rho_s$ and/or decreasing the annihilation rate constant $K_2$. Figure 9.9 shows the effect of an extreme 10-fold increase in $\rho_s$ to $10^{13}$, reduction in $K_2$ coupled with a reduction in the rate of K evaporation, $g$. A full list of parameter values is recorded in the "surface weighted KBr" column of table 9.4. As figure 9.9b shows, virtually all F centres
produced now decay at the surface freeing potassium atoms, but the formation of a potassium overlayer is still very slow only reaching 1% after two hours. In fact the degree of surface metallisation the model is able to predict is worryingly low, significantly below the 8% caesium excess Daengendorf et al. (1990) measured after 10 hours UV irradiation with a much lower flux (10^9 photons/mm^2/s compared to 5 x 10^10 used in the model).

The most conclusive evidence that it is F centres and not surface metallisation that causes the bulk of quantum efficiency degradation comes from the shape of the two curves in figure 9.9a, F centre concentration shows the characteristic rapid growth in concentration followed by a levelling off which if inverted would be very similar to the photoyield decay curves seen in chapter 8. Inverting the surface metallisation curve, by contrast, would predict an almost linear quantum efficiency decay. This is better illustrated by figure 9.10, the predicted photoyield variation over two hours irradiation. The top curve represents how the photoyield would be reduced solely as a result of surface metallisation, the lower curve solely by F centres trapping electrons while the triangles represent the combined effect of both these mechanisms. Also plotted (squares) is the measured photoyield degradation for KBr at 2keV, the parameters for which were used in the model. The overall comparison between theory and experiment is highly encouraging. The absolute degree of quantum efficiency degradation is dependent on a wealth of unknowns, mainly the percentage of vacant F centres and the true electron cross section of F centres at room temperature. Slightly more worryingly is the fact that the measured efficiency decay, after the rapid initial damage has been inflicted, is more pronounced than predicted. This could be the result of other defect species trapping electrons but is most likely because the model is under-estimating the surface metallisation, a modelling solution to which is unclear but probably relies on either a true defect diffusion model or a more accurate determination of the defect diffusion and interaction parameters. That said, the general agreement in curve shapes combined with the scope to match the quantum efficiency degradations observed gives one confidence that it is in fact F centre trapping that is the main cause of photoyield reduction in alkali halide detectors.

As the absolute magnitude of photoyield degradation depends on a large number of unknowns trying to fit possible parameter values to the data presented in Chapter 8 is not a terribly valuable exercise. However, it would be interesting to test the effect on the relative predicted degradation that changing one of the experimental parameters might have. The independence
Figure 9.9: a) Defect concentrations and fraction of potassium surface coverage under extreme surface dominant parameters (column 2 in table 9.4). b) Comparison of formation, annihilation and trapping rates for F centres demonstrating the lack of scope for further significant enhancement of surface metallisation for flux rates used in our synchrotron experiments.
Figure 9.10: Relative quantum efficiency degradation of KBr with 2keV photons normally incident showing the small linear effect caused by surface metallisation and the large exponential like effect due to F centre trapping. The combined model and experimental data are also plotted for comparison, note the absolute magnitude of the degradation is highly dependent on the assumed values of a number of unknowns.
on the rate of flux accumulation as mentioned above was probably the most unexpected result of our photoyield measurements. Figure 9.11a displays the predicted photoyield degradation for four orders of magnitude variation in the flux rate which fails to really solve the mystery. Each 10-fold increase in flux leads to a factor of two decrease in the degraded photoyield, the other factor of five being taken up by the increase in defect annihilation and trapping. Figure 9.11b compares F centre creation and annihilation/trapping rates for relative flux rates denoted 10 and 100 in figure 9.11a. The difference between the F centre production rate and the combined destruction processes (i.e. the clear area beneath the F centre production rate) can be seen to be relatively much smaller at the higher flux rate, but as the y axis scale is a factor of ten greater we get a factor of 2 more F centres.

The angular variation in rate of photoyield degradation is hard to test using this model as for grazing angles electron escape will be through regions where F centres are not created (unshaded regions in figure 9.6) and a full diffusion model would be required to estimate the F centre concentrations here. The main effect of photon energy will be on absorption, with at higher energies less photons being absorbed in given region, which is somewhat compensated by more F centres being created per photon. Calculations of the magnitude of these effects were made when constructing table 9.3 which clearly shows for our CsI synchrotron measurements, the rate of F-H pair production is virtually identical at 2.5 and 3.5 keV. However, the actual measurements presented in the preceding chapter show photon energy to be a determining factor as to the rate of degradation, with lower photon energies causing greater damage. These plots (figures 8.6 and 8.7) do though examine the efficiency variation with accumulated dose, against the energy dependence is a factor of 2 less. Also the distribution of F centre production with depth will vary with energy, but the model cannot account for this. Accounting for both these factors the discrepancy probably no longer exists.

Figure 9.12 shows the predicted CsI photoyield behaviour while the beam was switched off and then on again. Without continual F creation, the model processes of diffusion, annihilation and trapping are all still valid. The full curve represents the potential efficiency of the photocathode, added to that are sample predicted values while the beam was on and this measurements could be taken. This is done to make comparison with real data, as plotted in the previous chapter, easier. With the beam off, we have little idea of the shape of the recovery phase as no measurements can be taken, but the magnitude of the first recovery is reasonable. A number of
Figure 9.11: Upper) Predicted degree of CsI quantum efficiency degradation for 3 orders of magnitude variation in X-ray flux. Curves marked 1 and 100 representing the beamline 3.4 flux at 3610eV and 2000eV respectively for comparison with figure 8.10. Lower) The fate of F centres produced for relative intensities of 10 and 100 in the top figure. The unshaded area below the F centre creation rate represents surviving F centres capable of trapping photoelectrons.
Figure 9.12: Predicted quantum efficiency degradation and recovery characteristics of a CsI photocathode under synchrotron level doses.

data sets do show the second recovery to be smaller than the first, but a number do not so this isn’t too worrying. The shapes of the decays with beam on is again similar to the experimental data with the two component nature of the decay visible.

Finally, what does the model have to say for the various possible explanations for the radiation softness of thallium doped CsI as seen in figure 8.12? The two suggestions made in section 9.8 both fail to predict the behaviour of figure 8.12. Obviously the theory centred on thallium atoms anchoring the caesium metal islands has no validity if the surface metallisation is so small as to hardly affect the photoyield. That thallium atoms act as H centre traps increasing the concentration of F centres as less are annihilated also looks unlikely after viewing figure 9.13. Although increasing the impurity concentration (counting Tl as an impurity of this type) can be seen to reduce the radiation hardness of photocathode, the curve shapes became increasing contorted, while the experimental data shown Tl doping simply increased the degree of degradation without affecting the curve shape.
Figure 9.13: Predicted degree of photoyield degradation for different levels of H centre trapping impurities. The mole fraction of Ti in our doped sample was $5 \times 10^{-4}$.

9.10 Comparison with Scintillation Measurements

Adapting the model to predict variations in scintillation light yields has not been attempted but given the available knowledge of the affect of colour centres on scintillation light we can use the model to shed some light on the scintillation measurements displayed in figure 8.13. The variation of scintillation yield under X-ray illumination presented in this figure exhibited three interesting properties,

1. An initial rapid rise in scintillation yield of approximately 5%.
2. The yield peaking and then tailing off slowly by up to 8% after 50 minutes.
3. Blocking the source for 30 minutes led to a 5-10% yield reduction from which the sample never recovered.

Of these, ii) the reduction in scintillation yield, should be the easiest to explain. All colour centres are known to absorb light at various visible wavelengths. In fact, that is why they were
so named. Woody *et al.* (1992) examined the change in transmission of a thallium doped CsI crystal under gamma irradiation and found significant reductions especially between 400nm and 500nm where the peak response of the combined scintillator and photomultiplier tube system occurs. The problem with this explanation is that while the effect is significant for scintillator crystals, coated photocathodes are unlikely to suffer serious performance degradation from falls in transmissivity. Also the shape of the light yield decay is linear unlike the growth curves for any major defect species, in fact the most linear growth curve is that for surface metallisation suggesting internal reflection from a partially metallised surface may be the dominant effect. The shape of the initial rise in yield most closely follows the F centre concentration. As well as reducing transmissivity, F centres in the concentrations predicted have the potential to shift the emitted wavelength peak of the scintillation material toward that of the PMT peak response in much the same way adding dopants does, but only an experimental examination of the emitted wavelength can truly determine if this is what is occurring in our samples. It would though also explain the third result above, why blocking the source for 30 minutes caused a reduction in light yield when re-exposed to X-rays. During those 30 minutes the F centre concentration would have fallen significantly, but as many of them would have interacted with the surface, the surface metallisation would have dropped much less. Unblocking the X-ray source the yield would be lower, and by the time the F centre concentration was back up again the degree of surface metallisation would be far higher, so the yield could never recover to its original level.

### 9.11 Conclusions

The model described in this chapter provides insight into how well-known atomic defects produced in alkali halides cause the quantum efficiency degradations observed in both KBr and CsI photoconverters and scintillators. This, as the first study of its type, provides a basis for a thorough experimental analysis of the problems likely to be encountered using alkali halide detectors in high flux environments and how varying experimental parameters could improve radiation hardness, improving the lifetime and increasing the possible uses of such detectors. With

---

*The peak light output from CsI(Tl) is around 600 nm while the PMT is most efficient detecting light of wavelength just above 300nm. Data from Scionix Scintillation Detectors catalogue by P. Schotanus. Scionix Holland BV, PO Box 143, 3980 CC Bunnik, The Netherlands.*
F centre production the major villain, two obvious ways to reduce their concentrations would be to use ultra pure samples. The samples used during the experiments described in the preceding chapter were only 99.9% pure. In an ultra pure sample very few H centres would be trapped at impurities leaving more free to annihilate F centres. Operation at a higher temperature where F centres are also more mobile and more likely to interact with the surface would also help and bring the additional advantage of increasing desorption from any alkali metal overlayer. Another possibility would be regular illumination with F band while the photocathode was in a halogen environment to see if any of the halogen atoms would be taken up by the sample.

Before improving and using the model to discover if there are other ways to reduce or characterise radiation degradation, one must ask how confident we can be in its conclusions. There are a huge number of factors in table 9.4 which are unknown, as is the f centre electron cross section at room temperature and the number of likely free f centres. The main defect diffusion and interaction cross sections with one another are not too important, as long as we know which interactions are possible and which dominate. The comparative differences in the likelihood of a given interaction in NaCl, KBr or CsI is unlikely to be that great and so even if we do not know the magnitude of F-H annihilation, F centre trapping at F, M or R centres etc one can be confident that annihilation dominates, followed by trapping at F centres. Not knowing how many F centres are annihilated means we cannot know the absolute degree of quantum efficiency degradation and this is why I have not fixed it so as to agree with a couple of data sets, any such comparison would be meaningless. Knowing the dominant processes, however, means we know which new centres are most likely to be created and with it how in time a complex balance between defects is reached. Thus, we know the time histories of defect concentrations and it is the shape of these curves that has almost exclusively been used to determine which defect species or surface metallisation has resulted in which part of the photovield or scintillation yield degradation curve. Also Levy (1991) and others have measured the growth curves of various of the defect species and found them well fit by both the model’s predictions and for F centres a sum of exponentials similar to that fitted to the efficiency degradation curves in the preceding chapter.

The important unknown is the number of free F centres, that is F centres that have not trapped a second electron. The model assumes this to be constant given that F centres are continually

---

6The F band is the region of the visible spectrum strongly absorbed by F centres.
created and the number of electrons to be trapped by them is large, but if this is not so, the curve shapes on which most of the above analysis is based changes. How any defect model and the photoyield degradations can then be matched is not known by the author, and given the two should match hopefully the assumption is valid. Confidence can be taken from the fact that much of the data, including behaviour while the samples are not exposed to beam, is predictable by the model with reasonable and self consistent parameters.

9.12 Summary: Radiation Damage

This research has surely raised more questions than it has answered, but from both practical and theoretical standpoints continuing this research is worthwhile. Experimentally, characterising the radiation damage of alkali halides proved deceptively difficult. A method of ensuring the similarity of samples pre irradiation would have been the most useful addition.

The most surprising results are surely those varying the flux. These should be repeated with a range of filters blocking the beam so other parameters remain identical. Finally decay during the first 30 seconds data accumulation proved significant, but without knowing when during those 30 seconds the beam shutters were opened these early data was invalidated. A mechanical method of coinciding the start of measurements and the opening of the shutter would have helped significantly in our data analysis.

The model presented in this chapter shows the main causes of degradation to be i) F centres and ii) surface metallisation. Knowing this future modelling can concentrate on introducing physically accurate second order diffusion terms for the common defect species [f and h centres] and electrons while ignoring or adding minor corrections for the rarer ones. Lastly if the extended Jain Lidiard model is maintained, obtaining experimental parameter values for CsI and KBr is vital to ensure ones other efforts are worthwhile.
Chapter 10

Conclusions and Future Work

The work in this thesis has reported advances in the physical understanding of radiation damage processes in alkali halide detectors, the ending of the debate over the existence of the X-ray vectorial effect, and produced a new polarimeter design with many potential benefits over those currently available. Novel work on the use of synchrotron sources for the calibration of current and future astronomical X-ray spectrometers is also presented with the first broad band efficiency calibration of a pulse-counting microchannel plate.

10.1 Polarimetry

If we recall the aims of the polarimetry research program set out in Chapter 3, namely the production of a detector able at least, to determine the degree of modulation from bright active galactic nuclei (AGN), we can now see how the current technology shapes up. Unfortunately the experimental results obtained in Chapter 4 discount the most promising detection method, the so called X-ray vectorial effect which relies on polarisation sensitivity in the direction of emission of the primary photoelectrons being carried to secondaries which escape the photocathode’s surface. This highly accurate and ingenious experiment, relying as it did on the properties of the undulator source to ensure spurious modulation had not crept into the experiment, showed earlier experimental techniques, such as those described in Chapter 3, were too inaccurate and the modulations observed were a result of angular misalignments rather than a physical effect. The
Moderate modulations measured below 160eV in Chapter 4 were found to be well described by differences in the Fresnel reflectivities of light from different polarisation states, which, although important to consider when using any EUV grazing incidence detector or reflector, is not large enough to exploit in an astronomical polarimeter.

The model described in Chapter 5 was developed following an experiment by Shaw et al. (1994) which reported modulations up to 40% in the primary photoyield. The theoretical model, based on the electron transport properties of silicon confirmed these high modulations and led to the design of a new X-ray polarimeter exploiting this physical effect. By altering various parameters of the detector, namely the thickness, X-ray energy and voltage to discriminate against secondary electrons and polarisation insensitive Auger electrons the figure of merit for this 'multi-foil' polarimeter was optimised and compared favourably with the Bragg and Thomson scattering polarimeters to be flown on the Spectrum X-Gamma satellite. The building of such a detector will, however, be harder and require, for the reasons stated in chapter 5, a telescope with AXAF like resolution (0.5 arcseconds) and the collecting area of the SODART mirror assembly (1200cm^2) to be competitive with SXRP. More promising is that by building the detector of a different material 1 the polarimeter could be optimised for any energy range. For instance a beryllium foil polarimeter could be expected to be most efficient between 0.1 and 1keV where cosmic sources emit far higher fluxes but both Thomson scatterers and Bragg crystals are highly inefficient. Also if the discriminator voltage could be varied in flight it would allow the source polarisation to be measured in a number of X-ray energy bands (see for instance figure 5.14).

Much work still needs conducting, to both verify the primary photoeffect model and in developing an optimised polarimeter. Theoretical and experimental work should be aimed at characterising electron transport in lower Z materials especially Be. Further experimental effort could then directed toward testing the current model's predicted efficiencies and polarisation sensitivity for the silicon foil polarimeter. Given the difficulty we had in very accurately aligning one photocathode in a polarised X-ray beam, it would be difficult to be confident of the perfect alignment of many thin films without the checks a variable polarisation source provides. Therefore a better alternative would be to work with low Z materials in the energy range obtainable by the first

1The model is based on a silicon detector because the Shaw et al. (1994) experiment used silicon and because the electron transport properties of Si are well known. It is likely for reasons described in Chapter 5 that a lower Z material would be optimal.
harm oni of undulator emission on SRS beamline 5 and, as in Chapter 4, use the undulator as a fully and semi polarised source.

10.2 Broad Band MCP Quantum Efficiency determinations

The broad band alkali halide quantum efficiency determinations of Chapter 7 and more especially the bare microchannel plate (MCP) calibrations of Chapter 6 have demonstrated the vital role the <1eV energy resolution of synchrotron beamlines may play in the calibration of spectroscopic X-ray instruments. Our data show that using only available emission line energies and assuming simple steps at material absorption edges would have introduced an unacceptably large degree of spurious structure into future cosmic source spectra. Operation of the synchrotron with very low ring currents is shown to be feasible yielding usable flux levels for microchannel plate detectors. Methods to calibrate the responses were developed, given the absence of reference counters able to operate at low fluxes. Both detection efficiency and the strengths of the various absorption edges were shown to be strongly dependent on angle of incidence, as penetration through different thicknesses of the channel walls changes the constitution of the MCP glass where the photon is absorbed, demonstrating the importance of carrying out calibrations at the angle of incidence at which the final detector will be used. A number of issues concerning the accuracy of correcting the detected counts for the beam flux were addressed, as the results suggested the beamline characteristics for normal ring currents did not apply for our mode of operation. The conclusion reached was that problems with the microchannel plate were to blame for this discrepancy rather than with the calibration method, which has since been successfully utilised for a similar measurement series using coated MCPs.

The calibration of the AXAF CsI-coated HRC-S spectroscopic grating readout will be completed by similar relative quantum efficiency measurements throughout the 1-10keV range of the instrument. The absolute quantum efficiency of flight plates at various X-ray line energies will then be determined and the synchrotron calibration scaled so as to match these points.
10.3 Radiation Damage

The measurements described in Chapter 8 demonstrate that the problem of radiation damage in alkali halides is highly significant at synchrotron fluxes regardless of precise experimental parameters. For CsI a typical 90 minute synchrotron exposure (70 x 10^{12} photons or 7.8 Mrads) was shown to lead to a 50% degradation in quantum efficiency. With respect to accumulated flux, the decay in efficiency exhibited a marked "two component" nature, dropping rapidly at first, then steadying to an almost linear decay. Removing the sample from the beam led to a short lived recovery in quantum efficiency in almost all cases. One of the greatest experimental concerns was the degree of variability between measurements. Each measurement was conducted for both S and P polarised beams, but measurements rarely agreed and there was no consistency as to whether the S or P state was radiation hardest. With this degree of inaccuracy it is risky to draw too much from the data, but there were some interesting results. Lower energy X-rays caused the greatest degree of degradation, presumably as they are absorbed near the surface. Doping CsI with Ti also led to greater decays, but probably the most surprising result was that increasing the flux had virtually no effect on the rate of degradation.

Chapter 9 described the first microscopically-based model to examine the physical processes of defect production, diffusion, annihilation and trapping that result in the observed degradation in both quantum efficiency and scintillation yield. Of the various defect species and formation of surface over layers, evidence is presented that the dominant process in photoyield degradation is electron trapping at F centres. In the case of scintillators the model strongly suggests that F centres lead to an increase in the observed light yield as recorded by a PMT or maybe it is that F centres shift the wavelength of scintillation light such that it is more efficiently detected. But this increase in yield is countered by a second effect, possibly internal reflection at the metal rich surface, which soon reduces light yields again.

The defect model contains a large number of unknown parameters for which it would be more satisfactory to have better determined values (at present best fit values for NaCl are generally used, although as section 9.11 argues the conclusions above are not really reliant on the accurate determination of all these parameters). More important is to gain an understanding of the process by which F centres act as electron traps, to determine what are their cross sections at room temperature, how long is the extra electron held and is escape so probable that F centres
can trap large numbers of photoelectrons. Experiments where F centres are grown or inserted into crystals rather than X-ray induced, would avoid contamination of the system by so many defect species. If F centre trapping were found to be very similar to that described by Chapter 9 one could have even more faith in the model’s conclusions. Our ultimate aim of this work was to find a way of reducing the degree of damage or at least characterising it. Although ideas can be tested using the model the most important experiment work needs focusing on why measurements are so inconsistent and how this might be improved by more careful sample preparation, handling and experimental techniques.
References


Fraser, G. W., 1992. In: Photon detectors for Space Instrumentation, ESA SP-356, p. 97, ESA.


Jing, T., Goodman, C. A., Drewery, J., Cho, G., Wong, W., Lee, H., Kaplan, S. N., Perez-


Kifune, T., 1972.


195


Ultraviolet Spectroscopy and Polarimetry, 2283, 119, Proc. SPIE.


