Stokes parameter studies of electron-rare gas collisions.

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STOKES PARAMETER STUDIES OF ELECTRON-RARE GAS COLLISIONS

by

John Jay Corr

A Dissertation
submitted to the
Faculty of Graduate Studies and Research
through the Department of
Physics in Partial Fulfillment
of the requirements for the Degree
of Doctor of Philosophy at the
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ABSTRACT

STOKES PARAMETER STUDIES OF ELECTRON-RARE GAS COLLISIONS

by

John Jay Corr

Electron impact excitation of the rare gases has been studied via a series of polarisation correlation Stokes parameter measurements performed on an electron-photon coincidence apparatus. The Stokes parameters yield fundamental information on the magnitudes and relative phases of the scattering amplitudes which describe the scattering process, and also yield basic information about the elements of the density matrix describing the excited atomic state.

Use of an in-plane polarisation analyser allowed a detailed study of the relative so-called "height" of the excited state charge cloud immediately following the electron-atom collision. A breakdown of the reflection symmetry, with respect to the scattering plane, in the excitation process can be caused by a spin flip of the exciting electron, and would give a non-zero height parameter. Hence measurements of the charge cloud height parameter allowed a detailed analysis of the influence of spin in the excitation process.

Of critical importance to the performance of these measurements and the proper analysis of the experimental results is the definition of the scattering plane. Since the finite volume of the interaction region formed by the intersection of electron and gas beams can have depolarising effects, models which numerically simulate these effects and those due to the finite acceptance angles of the detectors have been developed. It was found that these "finite volume" effects were significant, under realistic experimental parameters, only at small angles (<5°) and at angles where the P₁ parameter approaches -1.
The atomic species studied were Ne, Ar, Kr, and Xe for electron scattering angles up to 50° and impact energies between 30 and 80 eV. Kr and Xe targets have naturally occurring isotopes with non-zero nuclear spin and in both cases the natural lifetimes of the excited states are long enough for complete hyperfine relaxation prior to photon decay. This causes depolarisation which must be taken into account.

After carefully taking into account these two depolarising effects very good agreement was found with theoretical predictions, in particular with distorted-wave Born approximation calculations. Within experimental error the height parameter was found to be zero and hence no evidence for spin flip of the exciting electron was found. This resolves the previous disagreement between a number of experimental groups and the theoretical predictions.
DEDICATION

This work is dedicated to my wife, Glenda, and son, Aaron.
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It is a great pleasure to acknowledge the endless guidance, patience, and enthusiasm of my supervisor, Dr. J. W. McConkey. He first aroused my interest in physics and introduced me to the world of scientific research. I also thank the faculty and staff of the Physics Department of the University of Windsor for creating a fine environment in which to learn.

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Finally, it is with deep appreciation that I thank my wife, Glenda, for her constant support and never ending patience over the past nine years. It was her loving encouragement which was truly responsible for the completion of this work.
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Chapter I
INTRODUCTION

The interaction of electrons with free atoms has been of scientific interest for a number of years, but until about 20 years ago a great deal of the experimental work was limited to the measurement of total and differential cross sections or to measurements of the intensity and polarisation of radiation emitted in the decay of excited states. Unfortunately all of these measurements provide only a "gross structure" of the excitation process.

Total electron impact excitation cross sections, by definition, contain an integration over all directions of the scattered electrons and simply assimilate into the measurement the fact that usually the greatest contributions to the cross section come from electrons scattered in the forward directions. It is then possible to have good agreement between theory and experiment for the total cross section for a particular inelastic process while the theoretical predictions for the large angle behaviour of the differential cross section may be completely incorrect. Differential electron impact excitation cross sections, by their nature, provide a more sensitive test between theory and experiment but do not distinguish between excitation to different magnetic sub-levels and hence fundamental information such as magnitudes and relative phases of the excitation amplitudes for the different magnetic sub-levels is not available.

Polarisation of the light emitted in the radiative decay of the excited atomic state is due to the generally anisotropic nature of the excitation process, in which different magnetic sub-levels are unequally populated. Its measurement then yields values for relative magnetic sub-level excitation populations, but this is again a value which is averaged over all electron scattering angles (and is related to the total cross section) and is an inevitable consequence of detection of radiation without regard to the scattered electron.
A technique which permits analysis of the light emitted from atoms excited by electrons scattered through a particular scattering angle is the electron-photon coincidence experiment. The basic arrangement consists of crossing an electron beam of fixed energy with an atomic gas beam. Electrons with an energy loss corresponding to the excitation energy of the state of interest are detected in delayed coincidence with photons resulting from the radiative decay of the excited atomic state. Because this technique does not involve averaging over magnetic sublevels or integrating over electron scattering angle, previously unavailable information is accessible with this method. Typically, one of two different measurements is made on the coincident radiation: (1) an angular correlation measurement, in which the angular distribution of the light is measured, or (2) a polarisation correlation measurement, in which the polarisation of the light is measured and the Stokes parameters determined. From either of these measurements it is possible to completely describe the excited atomic states and hence obtain information on the scattering amplitudes describing the collisional excitation.

Excited atomic states are described by target parameters known as alignment and orientation, which are related to their multipole moments. If reflection symmetry exists then the relative momentum of particles before and after the collision can be completely determined and the alignment and orientation of the target states immediately after the collision can be deduced. The atomic alignment is related to the shape of the excited state charge cloud and its alignment in space, while the atomic orientation is related to the angular momentum transferred to the atom during the collision. Since magnetic sublevel excitation is directly related to how angular momentum is transferred to the atom during the collision, determination of the alignment and orientation of the excited states gives the dynamics of the excitation process.
Under favourable circumstances electron-photon coincidence experiments yield a complete set of scattering amplitudes for the excitation process, including their relative phases. The favourable circumstances involve the coherence of the excitation process, in which the collision prepares an ensemble of atoms which can be represented as a linear superposition of a set of basis states. The magnitudes and phases of the coefficients in this expansion are the scattering amplitudes and are well-defined except for an overall phase factor. Electron-photon coincidence experiments measure interference terms between these excitation amplitudes and hence give a measure of the coherent nature of the excitation. When the excited atoms can be represented as a pure state the scattering amplitudes can be completely determined.

The fact that electron-photon coincidence experiments reveal much deeper insights into the electron-atom collision process is not without a price. Coincident signals are typically quite weak, meaning that in order to obtain measurements of reasonable statistical accuracy data collection times of up to several weeks for a single measurement are often required.

Polarisation correlation experiments must be performed taking into account two important depolarising effects: (1) the finite volume of the interaction region, and (2) internal hyperfine interactions for those species with isotopes of non-zero nuclear spin. The electron-atom scattering takes place in a small region formed by the intersection of the electron beam and the atomic gas beam. Although the dimensions of the interaction region are finite, they are usually much smaller than the distances from the interaction region to the photon detector and to the entrance aperture of the electron energy-analyser. For this reason it is usually assumed that the finite volume of the interaction region does not affect the signal obtained by the photon detector and the electron energy-analyser, but this can be an incorrect assumption.
In this work the electron-photon coincidence technique has been used to study electron impact excitation of the rare gases Ne, Ar, Kr, and Xe for electron scattering angles up to 50° and for electron impact energies between 30 and 80 eV. The Stokes parameters \( P_1, P_2, P_3, \) and \( P_4 \) have been measured and various quantities evaluated, including the charge cloud height parameter \( \rho_{oo} \). Analysis of the measured quantities incorporated the depolarising effects of the finite volume of the interaction region and of internal hyperfine interactions for those species with isotopes of non-zero nuclear spin. Effects due to the finite volume of the interaction region were simulated using an analytical model as well as a strictly numerical model. It was found that although the finite volume of the interaction region had greatest affect on \( P_4 \), which is a coincident polarisation measurement in the scattering plane formed by the incident electron beam and the scattered electron, this effect was usually small compared to that of hyperfine structure depolarisation, whenever it was involved. Non-zero values of the height parameter \( \rho_{oo} \) can indicate electron spin flip, but no experimental evidence for such a process was found. This resolved a significant disagreement between experiment and theory which had existed following publication of some Stokes parameter data by Plessis et al (1988) in which no account of the depolarising effects due to the finite volume of the interaction region, or to the hyperfine interaction, had been taken.
Chapter II
REVIEW OF PREVIOUS WORK

Since the electron-photon coincidence technique permits obtaining the most
detailed and fundamental information about the excited atomic states formed by
collisions of electrons and atoms there has been a vast body of work, both experimental
and theoretical, performed in this field. This work has been extensively reviewed and
discussed (Blum and Kleinpoppen 1979, McConkey 1979, Hanne 1983, Slevin 1984,
Andersen et al 1986, Slevin and Chwirot 1990) with one excellent review (Andersen
et al 1988) presenting the largest compilation of previous studies. The latter authors
present a valuable, comprehensive analysis of the varied parametrisation schemes for
describing the excitation process.

Macek and Jaecks (1971) presented the first detailed basic theory of electron-
photon coincidence measurements and the pioneering experiment was an angular
correlation study by Eminyan et al (1973). Since then a number of atomic targets have
been studied with the 2P excitation of helium being the most widely investigated. The
reason for this is that the He situation is relatively simple since LS coupling holds strictly
and spin-orbit interactions during the collision can be neglected. The justification for
this assumption is the Percival-Seaton hypothesis (Percival and Seaton 1958) which,
in essence, states that the collision interaction time (∼10⁻¹⁵ sec) is much shorter than
any spin precession time due to spin-orbit interaction (∼10⁻¹² sec) meaning that the
electron spin is fixed in space during the collisional interaction. The spin of the system
and its orientation in space is thus conserved during the collision, allowing electron
spin to be factored out of the problem, and thus the positive reflection symmetry (with
respect to the scattering plane) of the atomic wavefunction is conserved during the
collision. Figure 1 illustrates the concept of reflection symmetry with respect to the
scattering plane (the x-z plane in this diagram). Clearly the angular dependence of the

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$| \rho_x \rangle$ and $| \rho_z \rangle$ orbitals possess positive reflection symmetry while the $| \rho_y \rangle$ orbital displays negative reflection symmetry with respect to the scattering plane. If a dipole oscillator perpendicular to the scattering plane is excited it must be observed from a direction other than perpendicular to the scattering plane (hence the in-plane $P_\theta$ measurements). Measurement of the polarisation of its radiation allows the height parameter $\rho_{00}$ to be determined and thus a further simplification of the analysis.

![Reflection symmetry with respect to scattering plane](image)

**Figure 1. Reflection symmetry with respect to the scattering plane.** Angular dependence of $| \rho_x \rangle$, $| \rho_y \rangle$ and $| \rho_z \rangle$ orbitals illustrating symmetry properties with respect to the scattering plane. Taken from Slevin 1984.

He excitation is described by a scattering amplitude which may be decomposed into a direct part and an exchange part. These two amplitudes are in principle experimentally indistinguishable and are independent of the original orientation of the electron spin. The initial state is incoherent due to lack of spin polarisation of the incident electron beam, but since spin can be factored out the process can be treated as a fully coherent case and hence the excited state may be described by a single pure state. Adding to the convenience of studying He is the fact that there are no hyperfine
structure effects to contend with, and it is very easy to handle experimentally. Standage and Kleinpoppen (1976) published the first complete analysis of electron impact excitation of the 2'P and 3'P states of He.

After He the next most widely studied species has been atomic hydrogen, which represents an increase in the level of complexity since an atomic doublet state is excited and therefore a different analysis and interpretation of the experiment is required. For the same reasons as in the He case an LS coupling scheme can be adopted for the collision process in which the total spin S of the combined system of the scattered electron and the atomic electron is conserved, and thus the atomic electron spin is assumed to be fully decoupled from the orbital motion. The direct and exchange amplitudes are now composed of combinations of the singlet and triplet scattering amplitudes and these amplitudes offer two different processes over which the experiment averages. Hence, in contrast to the He case, the excited state can no longer be described by a single pure state and thus the process is only partially coherent. Since experimental preparation of H atoms involves dissociation of H₂ the target density is generally rather low and hence the vast majority of the work on H has been theoretical. Nevertheless the first experimental measurements reported by Dixon et al (1978) and other following work has provided a challenging set of data for testing of electron scattering theories.

The next level of complexity involves heavier atoms, in which LS coupling is no longer strictly valid since now total electron angular momentum J, rather than simply the electron orbital angular momentum L, is the relevant quantum number. Electron spin may play a role in the excitation either through spin flip (because spin-orbit effects might be very strong) or exchange with target electrons. This statement is equivalent to saying that the atomic reflection symmetry may change during the collision and states having negative reflection symmetry with respect to the scattering plane may also be
excited. More generally, it can be said that the collisionally excited charge cloud may have a finite height (i.e. $\rho_{00} \neq 0$), or in classical terms, the excitation of an oscillator perpendicular to the scattering plane may be occurring. The remainder of this chapter will focus on previous work done on the heavier rare gases and on effects due to the finite volume of the interaction region.

Transitions of interest in the heavier rare gases are from an isotropic $^1S_0$ ground state to $ns'[1/2]^0$ or $ns'[3/2]^0$ excited states (Racah notation). The electron charge cloud is now determined by the unoccupied p-orbital of the atomic core and the total angular momentum of the excited state is $J = 1$, the fine structure being completely resolved. Spin flip due to spin-orbit interaction and due to exchange cannot be strictly separated and the excited states are described as mixtures of $^1P_1$ and $^3P_1$ states. Since electrons of different spin may have different excitation amplitudes they contribute incoherently to the excited atomic state so that full coherence can no longer be assumed. Since the total polarisation parameter $P^*$ (see Chapter III for definitions) is a measure of the coherence of the excitation the difference in spin-up and spin-down amplitudes will give the possibility of $P^* < 1$. The height parameter $\rho_{00}$ gives the relative probability for spin flip perpendicular to the scattering plane.

Ninety-five percent of all electron-photon coincidence experiments are concerned with excited p-states, and the vast majority of these are angular correlation experiments because of their relative simplicity. A major drawback to these studies is that the sign of the angular momentum transfer during the collision cannot be determined, while it can be unambiguously determined in polarisation correlation experiments. The only reported polarisation correlation investigations of the heavy rare gases are those of the Windsor group (Malcolm and McConkey 1979, Khakoo and McConkey 1986, 1987 and Plessis et al 1988) and those of Martus and Becker (1989).
Khakoo and McConkey (1986, 1987) reported out-of-plane linear ($P_1$, $P_2$) and circular ($P_3$) polarisation correlation (Stokes parameter) results for Ne, Ar, and Kr. These measurements allowed a study of the part of problem which has positive reflection symmetry and the circular polarisation measurements enabled the sign of the angular momentum transfer in the collision to be unambiguously determined. For all cases studied the authors found that for electron scattering angles less than 30° the angular momentum transfer is positive and very similar (see also McConkey et al. 1988). Measured total polarisations of close to unity suggested that essentially full coherence of this part of the excitation was occurring.

Plessis et al. (1988) presented the first in-plane Stokes parameter measurements for Kr and Xe and found large deviations of their measured $P_4$ parameters from unity over a certain range of smaller electron scattering angles. This indicated that the excited state charge clouds possessed significant height perpendicular to the scattering plane and suggested that the resulting non-zero $\rho_{xx}$ values were due to a breakdown of the reflection symmetry, caused by a spin flip of the exciting electron.

Martus and Becker (1989) reported measurements of the $P_1$ coherence parameter for excitation of the "P₁" state of Ne and Ar by electrons which were scattered in the forward direction. For electron impact energies in the range 30 to 100 eV they measured $P_1 = +1$, which indicated, in agreement with the appropriate theories, that the excitation process proceeds via a pure transfer of orbital angular momentum, meaning that the process is the direct excitation of the LS-coupled singlet component of the excited state. The authors were careful to take effects of the angular resolution of the apparatus into account.

Neon is expected to resemble the He case somewhat closely since it is the lightest of the heavy rare gas atoms and hence should display the smallest effects of spin-orbit interaction. Balashov et al. (1981) performed the first theoretical calculation for Ne
using an eight-state multichannel diffraction approximation (MCDA), but they did not take spin-orbit effects into account and hence implicit in their calculations is $\rho_{oo} = 0$. A first-order many-body theory (FOMBT) calculation by Machado et al. (1982) considered different target couplings and was the state-of-the-art until the distorted-wave Born approximation (DWBA) calculations of Bartschat and Madison (1987). This latter work investigated the importance of relativistic spin-dependent effects both on the description of the Ne, Ar, Kr, and Xe target states and on the wavefunction of the continuum electron. For the smaller scattering angles corresponding to this work these effects were found to be small even for a heavy target such as Xe.

The first experimental study of Ne was the angular correlation effort reported by Ugbabe et al. (1977) in which they examined the $3s^1[1/2]_0^0 (3P_1)$ state for impact energies of 80 and 120 eV. Other than the usual problem of the inability to experimentally resolve the $1P$ and $3P$ states there is a flaw in their work in that they treat this case exactly as the He $1P_1$ case. This is not a valid treatment since it can no longer be assumed that $\rho_{oo} = 0$. Although Khakoo and McConkey (1986, 1987) could not resolve the singlet and triplet states they estimated contribution to their signal of less than 20% from the triplet state in their study of the $1P$ state. These two studies were in good agreement although the range of the electron scattering angles in common was very small. Agreement with the MCDA work of Balashov et al. (1981) and with the FOMBT calculations of Machado et al. (1982) is good out to a scattering angle of about 20°, beyond which the these theories seem to break down. Although the experiments seemed to agree slightly better with the DWBA theory of Bartschat and Madison (1987), the same disagreement at larger angles was present. Since Ugbabe et al. (1977) assumed $\rho_{oo} = 0$ and Khakoo and McConkey (1986, 1987) only measured out-of-plane Stokes parameters comparisons are limited to $P_1$ and $P_2$ parameters, though it is noted that the theoretical work reports $\rho_{oo}$ values very close to zero even beyond the angular
ranges covered by the experiments.

The $P_1$ results for forward scattered electrons reported by Martus and Becker (1989) appear to be in agreement with the other experimental efforts although none of these efforts actually report on forward scattering. As Martus and Becker (1989) note, their results are in good agreement with the DWBA calculations of Bartschat and Madison (1987).

In argon the mixing coefficients for the singlet and triplet states are closer to one another in absolute value than in the Ne case, but it is still possible to refer to a predominantly singlet state ($|s^{-1}1/2\rangle$) and triplet state ($\pm|s3/2\rangle$), though differences should be much less pronounced. Theoretical treatments of the MCDA data of Balashov et al (1981), the FOMBT calculations of da Paixao et al (1984), and the DWBA work of Bartschat and Madison (1987). Experimental angular correlation measurements have been reported by Arriola et al (1975), Malcolm and McConkey (1979), and Pochat et al (1980). Although these efforts combine to cover four different electron scattering angles, the largest is only 10°. Slevin and Farago (1975) have pointed out that Arriola et al (1975) were not able to experimentally separate the $^1P$ and $^3P$ states yet they incorrectly determined four independent parameters $\lambda_1$, $\lambda_3$, $\chi_1$, and $\chi_3$ when only two parameters, $\lambda$ and $\chi$, pertaining to the incoherent sum of p-states were obtainable. Nevertheless the three experiments are in good agreement and also agree well with the work of Khakoo and McConkey (1986, 1987). For the case of 50 eV impact energy electrons being forward scattered Malcolm and McConkey (1979) measured $P_1$ values that were essentially unity for the "triplet" state, but for the "singlet" state they measured $P_1 = 0.4$ while Martus and Becker (1989) obtained $P_1 = 1$. These $^1P$ results of Malcolm and McConkey (1979) suggest significant spin-orbit coupling interactions which are not expected and which disagree with the FOMBT and DWBA predictions. Other than this all the experimental data have better agreement.
with the DWBA calculations which indicates that relativistic effects should be included in the target and continuum wavefunctions. At the electron impact energies and scattering angles covered by the experimental work, theory and experiment both seem to indicate that $\rho_{00} = 0$. It is noted that, just as in the Ne case, both the FOMBT and the DWBA calculations indicate substantial non-zero $\rho_{00}$ values for the $^3P$ state at scattering angles larger than those covered in the angular correlation work. Although Khakoo and McConkey (1986, 1987) extended their measurements out to 30° they could not distinguish between the singlet and triplet states and did not make in-plane measurements which would allow the determination of $\rho_{00}$.

For the krypton case the singlet-triplet mixing coefficients are very similar for the $5s\ [1/2]^0$ and $5s\ [3/2]^0$ states. This means they cannot be labelled as being predominantly singlet or triplet and hence their respective orientation and alignment parameters are very similar for their electron impact excitation. The only theoretical calculations for Kr are the FOMBT calculations of Meneses et al (1985), which include spin-orbit coupling in the target, and the DWBA work of Bartschat and Madison (1987). The three sets of angular correlation data reported were those of McGregor et al (1982), King et al (1985), and Danjo et al (1985). Since McGregor et al (1982) only reported the $\lambda$ and $\chi$ parameters, Stokes parameters cannot be derived from their results and in particular $\rho_{00}$ cannot be evaluated, making comparison with the polarisation correlation studies of Khakoo and McConkey (1987) and Plessis et al (1988) impossible. Excluding the results of McGregor et al (1982) the other four experiments are in very good agreement (the polarisation studies are from the same group). All four are in quite reasonable agreement with the DWBA treatment except for the $P_1$ Stokes parameter and in that they indicate substantial non-zero $\rho_{00}$ values at small scattering angles, indicating significant height to the electron charge cloud. The data of Plessis et al (1988) possess much smaller error bars than that of the other groups and the data
of Danjo et al. (1985) indicate \( \rho_{oo} \) values much larger than the other groups.

As indicated earlier, the results of Plessis et al. (1988) showed large deviations of their measured \( P_* \) values from unity. The other experiments seemed to substantiate this observation, and since \( \rho_{oo} \) is calculated directly from \( P_* \) and \( P_* \), deviations of this quantity from zero appear. Although Plessis et al. (1988) suggested that their \( \rho_{oo} \) values were due to a breakdown of the reflection symmetry caused by a spin flip of the exciting electron such a process at these small scattering angles is unexpected on theoretical grounds. The reasoning is that small-angle scattering corresponds to large impact parameters where the influence of any explicit spin-dependent interactions should be very small. The deviation of \( P_* \) from unity definitely seemed to occur when the in-plane photon detector coincided with the charge cloud alignment angle, \( \gamma \). These large discrepancies caused consternation among both experimental and theoretical investigators. Very recently Murray et al. (1990) reported the results of an angular correlation study of Kr. Although they did not include \( P_* \) and \( \rho_{oo} \) findings they did note that \( P_* \) departed from unity at scattering angles up to 50° with a 15° value of -1 being the extreme. Because of this large departure from unity the authors did not calculate \( \rho_{oo} \) values, claiming that they would be suspect. As explanation for these low \( P_* \) values the authors suggested that perhaps poor definition of the scattering plane was the cause.

All experimental studies of xenon have focussed on the 6s[3/2] state. The angular correlation data consists of the single scattering angle report of McGregor et al. (1982) (at an electron impact energy of 80 eV) and the study of Nishimura et al. (1986). Measurements of Plessis et al. (1988) taken at an impact energy of 50 eV, while not in good agreement with those of Nishimura et al. (1986), show the same features. These are as in the Kr case only the \( P_* \) departures from unity are larger (values now approach zero), and \( \rho_{oo} \) values indicate even greater height to the charge cloud. The work of Nishimura et al. (1986) indicated lower \( P_* \) values, larger \( \rho_{oo} \) values, and pos-
sessed substantially larger error bars. As in the Kr case the experimental observations contradicted the predictions of the sole theoretical work, the DWBA work of Bartschat and Madison (1987).

Because experimental $P_\pi$ and $P_{\alpha\alpha}$ values differed so drastically from the theoretical predictions considerable activity ensued in a number of laboratories to see if these data could be accounted for by experimental effects, in particular those related to the finite volume of the interaction region. While repeating the superelastic scattering experiment of Register et al (1983), in which electrons were scattered from laser-excited $^{138}$Ba ($^3P_\pi$), Zetner et al (1989, 1990) found that under certain circumstances the finite volume of the interaction region can have a dramatic effect on the measured quantities. Since electron-photon coincidence experiments can be viewed as the time-inverse process of superelastic scattering experiments this work suggested that serious experimental effects of this type might be occurring in these experiments also. Zetner et al (1989, 1990) performed extensive experimental tests regarding their interaction region but were unable to eliminate unexpected asymmetries in their observed signals. Through model calculations they found that the finite volume of the interaction region can severely affect the observed superelastic scattering intensity, even at scattering angles substantially removed from 0°.

The finite volume of the interaction region causes a loss of definition of the scattering plane formed by the intersection of the incident electron beam and the scattered electron, particularly at small scattering angles. This is of critical importance since information concerned with spin-dependent processes is deduced from the measurements. These concerns have been discussed by a number of authors (Martus et al 1988, Martus and Becker 1989, Hanne 1989, McConkey et al 1989). Recently
Simon *et al* (1990) performed a small angle electron-photon coincidence experiment on electron impact excitation of Hg($^3P_1$) and were very careful to attempt to properly allow for these effects.

In all the previously published work on the heavy rare gases there was no consideration of hyperfine depolarising interactions and Martus and Becker (1989) presented the only work which took into account angular resolution depolarising effects.
Chapter III
THEORETICAL CONSIDERATIONS

3.1 Introduction

Since the electron-photon coincidence technique permits the most fundamental aspects of atom excitation due to electron impact to be probed the theoretical description of this technique has received extensive attention. As a result a number of different formalisms and parametrisation schemes have been developed. In this work the density matrix formalism (Blum 1981) and the Stokes parameter scheme (P.L.O.) are combined following the method of Andersen et al (1988) to provide a clear, concise description of the excitation in terms of the parameters \( \gamma, \ P_1, \ L_\perp \) and \( \rho_{00} \).

To set the stage for the remainder of the chapter the first topic of discussion is the choice of coordinate frames and basis sets followed by a description of the Stokes parameters and how they can be determined by polarisation measurements. The density matrix formalism is then presented in conjunction with the state multipole moments. Next, a theoretical description of hyperfine structure effects is presented followed by a section which relates the multipole moments, and hence the density matrix, to the Stokes parameters. The chapter concludes with a section specialising the theoretical framework to \( P \rightarrow S \) transitions and the frame independent parameters which describe the excited state charge cloud are derived.

The review of Andersen et al (1988) treats the entire field of electron-photon coincidence studies in such an excellent fashion that the remainder of this chapter is essentially a condensation of their work and is not intended to be presented as an original treatment.
3.2 Coordinate Frames and Basis Sets

In electron-photon coincidence experiments the excited state of the atom is described by a density matrix which is relevant to a particular coordinate frame. It is obviously preferred to have a frame-independent parametrisation of the quantities describing the excitation. Such a parametrisation has been developed by Andersen et al (1988) and is closely related to the inherent symmetries of the excitation.

Figure 2 is a schematic illustration of the angular part of a collisionally induced charge cloud in a p-state atom. The scattering plane is defined by the directions of the incoming ($\vec{k}_{in}$) and outgoing ($\vec{k}_{out}$) electron momentum vectors. The incoming and outgoing plane waves have positive reflection symmetry with respect to only this plane. This, combined with the fact that the Hamiltonian for the interaction has positive reflection symmetry demands that the reflection symmetry with respect to the scattering plane of the total wavefunction of the combined system of electron and atom is conserved during the collision. If the initial state distribution is spherically isotropic then the excited state charge cloud will also display reflection symmetry. The preceding statements are consequences of the parity invariance of the scattering process. Parity invariance of the excitation requires that the net orbital angular momentum transferred with respect to a coordinate axis $i = x, y$ or $z$ ($\Delta \vec{L}_i$) be such that $\Delta \vec{L}_i \cdot \vec{k}_{out} = \Delta \vec{L}_i \cdot \vec{k}_{in} = 0$. Clearly the angular momentum (of the relative motion of the particles) transferred must be perpendicular to the scattering plane and is thus labelled $L_\perp$.

Such a model of the atomic charge cloud allowed Andersen et al (1988) to fully characterise the excited state by four frame-independent quantities: the relative height ($h$) (or width ($w$)), length ($l$), and alignment angle ($\gamma$) of the charge cloud and by its inherent angular momentum $L_\perp$. These quantities are incorporated into the commonly used parameters $\gamma$, $P_i$, $L_\perp$ and $\rho_{00}$ and are displayed in Figure 2.
Figure 2. Charge cloud of collisionally induced p-state. The scattering plane is defined by the incoming $\vec{k}_{in}$ and outgoing $\vec{k}_{out}$ momentum vectors. The parameters $l$ (or $w$), $h$, $\gamma$ and $L_\perp$ fully describe the excited state. The coordinate frame is the natural frame. Taken from Andersen et al (1988).

The conventional frame used in scattering studies is the collision frame in which the direction of the incident electron beam is declared the quantisation axis (the $z'$ axis). Hermann and Hertel (1982) pointed out that the natural frame, in which the quantisation axis (the $z^*$ axis) is chosen perpendicular to the scattering plane, allows
the inherent symmetries of the scattering to be better utilised to obtain a clear picture of the collision dynamics (see Figure 2). Using the superscripts "n" to denote the natural frame and "c" to denote the collision frame, the coordinate frames are related through $x^n = z^c, z^n = y^c, y^n = x^c$. Another convenient coordinate frame is the atomic frame which is obtained from the natural frame by rotating through an angle $\gamma$ about $z^n = z^c$ so that $x^a$ parallels the major symmetry axis of the charge cloud (which is also a symmetry axis for the dipole radiation pattern). The natural frame is often referred to as the more suitable frame, but for calculations it is often more convenient to use the collision frame.

Figure 3 displays the two most convenient choices for a p-state basis set. The molecular basis set composed of the $\mid p_x \rangle, \mid p_y \rangle$ and $\mid p_z \rangle$ orbitals is also referred to as $\mid \sigma \rangle, \mid \pi \rangle$ and $\mid \pi' \rangle$, respectively, and is related to the atomic basis set, defined by the magnetic quantum numbers $M$, through

$$\mid p_x \rangle = -[\mid 1 \rangle - \mid -1 \rangle] / \sqrt{2}$$

$$\mid p_y \rangle = i[\mid 1 \rangle + \mid -1 \rangle] / \sqrt{2} \quad (3.2.1)$$

$$\mid p_z \rangle = \mid 0 \rangle$$

where $\mid p_M \rangle = \mid M \rangle$ (with $L = 1$ suppressed for p-state excitation) is used interchangeably for the atomic basis set. Reflection invariance, for p-states, is expressed by the elements of the density matrix in the collision frame satisfying

$$(L M' \mid p \mid L M) = (-)^{M-M'} (L-M' \mid p \mid L-M) \quad (3.2.2)$$

From this point forward the atomic basis set will be the implied basis set unless otherwise stated.
Figure 3. Two p-state basis sets in the natural frame. The upper row shows the atomic basis set $|p_M\rangle$ defined by the magnetic quantum numbers $M$ and the lower row shows the molecular basis set composed of the $|p_\gamma\rangle$, $|p_\gamma\rangle$ and $|p_z\rangle$ orbitals. Taken from Andersen et al (1988).

3.3 The Stokes Parameters

Light emitted in a particular direction, from any radiation source, can be fully characterised by its total intensity and by three quantities known as the relative Stokes parameters. The linear Stokes parameters, $P_1$ and $P_2$, describe the linear polarisations of the light:

$$P_1 = \frac{I(0^\circ) - I(90^\circ)}{I(0^\circ) + I(90^\circ)} \quad (3.3.1)$$

$$P_2 = \frac{I(45^\circ) - I(135^\circ)}{I(45^\circ) + I(135^\circ)} \quad (3.3.2)$$

where $I(\phi)$ is the intensity of light with polarisation vector in the direction $\phi$ with respect to a quantisation direction, in the present case that of the incident electron beam. The
third Stokes parameter is the circular polarisation

\[ P_3 = \frac{l_R - l_L}{l_R + l_L} \] (3.3.3)

where \( l_R \) and \( l_L \) are the right and left circularly polarised components, respectively, with the convention of classical optics that R (L) photons have helicity -1 (+1). In this work, and in most other coherence studies, \( P_1, P_2 \) and \( P_3 \) are measured for light propagating perpendicular to the scattering plane (see Figure 4) along the z-axis in the natural frame.

Measurements of the Stokes parameters for light emitted in a particular direction permit the determination of only three of the four independent parameters that fully characterise the excited state. A polarisation measurement from a different direction is required. The usual measurement is

\[ P_4 = \frac{l_\parallel - l_\perp}{l_\parallel + l_\perp} \] (3.3.4)

for light emitted in the scattering plane perpendicular to the incident electron beam, where \( l_\parallel \) and \( l_\perp \) are the intensities of light with polarisation vector parallel to and perpendicular to the scattering plane, respectively. When the term "Stokes parameters" is used in the current context it is usually meant to include \( P_4 \) (\( P_4 \) is a Stokes parameter measured from a different direction).

The linear polarisation measurements exploit the fact that radiation from the decay of different magnetic sublevels has different polarisation, as indicated in Figure 5. For \( J = 1 \rightarrow J = 0 \) transitions linear light polarisation parallel to the x-axis, when observed normal to the x-axis, is due to transitions from \( M_J = 0 \) while transitions from \( M_J = \pm 1 \) produce linear light polarised perpendicular to the x-axis. By observing photons with a particular one of these polarisations in coincidence with the exciting electrons the excitation of either the \( M_J = 0 \) or the \( M_J = \pm 1 \) sublevels can be selectively isolated,
though the $M_s = \pm 1$ sublevels are indistinguishable with this method.

King et al (1972) first illustrated the importance of $P_1$ measurements in coincidence with forward inelastically scattered electrons for $P \to S$ transitions. They demonstrated that it is possible to make "pseudo-threshold" polarisation measurements which obey the same selection rule (based on angular momentum arguments) as for excitation at threshold. If the axis of quantisation is taken along the direction of the incident electrons, the incident electrons have zero angular momentum about this axis and since the initial atomic state is an s-state the total component of the initial angular momentum is zero. At threshold the scattered electrons carry off very little energy and
Figure 5. Polarisations due to different magnetic sublevels. For observation perpendicular to the x-axis $M_I = 0$ decays emit light polarised parallel to the axis while $M_I = \pm 1$ decays emit light polarised perpendicular to the axis. Adapted from Goeke et al (1989).

Therefore no angular momentum and hence there can be no change in the component of the atomic orbital angular momentum. This argument results in the selection rule $\Delta M_I = 0$. McFarland and Mittleman (1968) pointed out that this argument stops short of considering the spin of the incident electron, but agree that at threshold the s-wave part of the emerging electron dominates and the usual practice of neglecting spin-dependent forces at threshold may be followed, giving the selection rule $\Delta M_I = 0$.

"Pseudo-threshold" measurements obey the same selection rule since only forward scattered electrons are considered and the incident and scattered electrons both move along the x-axis, meaning neither have angular momentum about that axis and therefore cannot change the component of the orbital angular momentum of the atom. Since excitation of $M_I = \pm 1$ sublevels from $M_I = 0$ can then only occur by transfer of spin angular momentum, and since, as discussed above, different magnetic sublevels
emit light of different polarisations, a measurement of $P_t$ is directly related to the relative $M_j = \pm 1$ and $M_f = 0$ populations. As mentioned earlier in the discussion of the experiment of Martus and Becker (1989), threshold $P_t$ values of $+1$ indicate direct excitation of the $^1P$ component while threshold $P_t = -1$ indicates pure exchange excitation of the $^3P$ component via spin transfer. A value of threshold $P_t$ between $-1$ and $+1$ reveals the relative contributions of the two excitation mechanisms at a particular incident energy.

3.4 State Multipole Moments and the Density Matrix

The theory of electron-photon coincidence experiments has appeared numerous times in the literature in various different forms. Hence no attempt to rigorously develop the treatment is given in this work, instead only the important results and the key steps leading to them are presented. The theoretical development here is taken from Andersen et al (1988).

The density matrix of a set of states characterised by the angular momentum quantum numbers $J M_j$ is denoted $\rho_{J', M', J M}$. The state multipole moments $\langle t (J' J) \rangle_{KQ}$ provide an irreducible representation of $\rho_{J', M', J M}$ in terms of the irreducible tensor operators $T (J' J)_{KQ}$. The advantage of this representation is that the various symmetries inherent in the excitation are exploited allowing separation of dynamical and geometrical factors in the collision process. Rotations between reference frames and projections of state distributions onto the different angular momentum coupling schemes also follow conveniently from this representation.

The state multipole moments as expressed by Blum (1981) are

$$\langle t (J' J) \rangle_{KQ} = \sum_{M' M} (-)^{J' - M'} (2K + 1)^{1/2} \left( \begin{array}{ccc} J' & J & K \\ M' & -M & Q \end{array} \right) \rho_{J', M', J M} \quad (3.4.1)$$
where $\langle J^\cdot J \rangle_{kQ}$ is the statistical tensor operator and the angular brackets indicate averaging over all spins. The density matrix elements follow from inverting this equation:

$$
\rho_{J^\cdot M^\cdot, J^\cdot M} = \sum_{kQ} (-1)^{J^\cdot M^\cdot} (2K + 1)^{1/2} \left( J^\cdot M^\cdot - J, M \right)_{Q} \langle J^\cdot J \rangle_{kQ} . \quad (3.1.2)
$$

In the literature excitation parameters are expressed in terms of the real multipole moments rather than the state multipole moments since they lead to a more compact form. The relations of the real multipole moments to the state multipole moments are given by

$$
\langle I' (J) \rangle_{kQ} = \frac{\langle I (J^\cdot J) \rangle_{kQ}}{I^{(k)}(J)} . \quad Q = 0 \quad (3.1.3)
$$

$$
\langle I' (J) \rangle_{kQ} = \frac{1}{\sqrt{2}} (-)^{Q} \langle I (J^\cdot J) \rangle_{kQ} + \langle I (J^\cdot J) \rangle_{k-Q} \frac{I^{(k)}(J)}{I^{(k)}(J)} . \quad Q > 0 \quad (3.1.4)
$$

$$
\langle I' (J) \rangle_{kQ} = -\frac{i}{\sqrt{2}} (-)^{Q} \langle I (J^\cdot J) \rangle_{kQ} - \langle I (J^\cdot J) \rangle_{k-Q} \frac{I^{(k)}(J)}{I^{(k)}(J)} . \quad Q < 0 \quad (3.1.5)
$$

with

$$
I^{(k)}(J) = \frac{2^{k}(2K + 1)^{3}}{K!} \left[ \frac{(2J - K)!}{(2J + K + 1)!} \right]^{1/2} (-)^{2J} . \quad (3.1.6)
$$

For the excitations of interest $J = 1$ and the $V^{(k)}(J = 1)$ values are

$$
I^{(0)}(1) = \frac{1}{\sqrt{3}} , \quad I^{(1)}(1) = \frac{1}{\sqrt{2}} , \quad I^{(2)}(1) = \frac{1}{\sqrt{6}} . \quad (3.1.7)
$$

Table 1 lists the relations between the collisionally induced real multipole moments and the density matrix elements in the natural frame using the atomic basis set for p-state ($J = 1$) excitation. $J = 1$ is suppressed in the notation.

As previously mentioned, this parametrisation in terms of the real multipole moments permits simplification in projecting state distributions from one angular
Table 1. Multipole moments and density matrix elements for $J = 1$. Relations between the collisionally induced multipole moments and the density matrix elements in the natural frame for the atomic basis set for $J = 1$. ($J = 1$ is suppressed in the notation.)

<table>
<thead>
<tr>
<th>Multipole Moments</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt;T_{00}&gt; = 1$</td>
</tr>
<tr>
<td>$&lt;T_{10}&gt; = \rho_{11} - \rho_{-1-1} = 2\rho_{11} - 1 + \rho_{00}$</td>
</tr>
<tr>
<td>$&lt;T_{20}&gt; = \rho_{11} - 2\rho_{00} + \rho_{-1-1} = 1 - 3\rho_{00}$</td>
</tr>
<tr>
<td>$&lt;T_{22}&gt; = 2\sqrt{3} Re(\rho_{1-1})$</td>
</tr>
<tr>
<td>$&lt;T_{22_-}&gt; = -2\sqrt{3} Im(\rho_{1-1})$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Density Matrix Elements</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho_{00} = \frac{1}{3}(1 - &lt;T_{20}&gt;)$</td>
</tr>
<tr>
<td>$\rho_{11} = \frac{1}{3} + \frac{1}{2} &lt;T_{10}&gt; + \frac{1}{6} &lt;T_{20}&gt;$</td>
</tr>
<tr>
<td>$\rho_{-1-1} = \frac{1}{3} - \frac{1}{2} &lt;T_{10}&gt; + \frac{1}{6} &lt;T_{20}&gt;$</td>
</tr>
<tr>
<td>$\rho_{1-1} = (2\sqrt{3})^{-1} (&lt;T_{22}&gt; - i&lt;T_{22_-}&gt;)$</td>
</tr>
</tbody>
</table>
momentum coupling scheme to another. The state distribution in the coupled system $(LS)JM_s$, where $S$ is the atomic electron spin and $J$ is the coupled angular momentum, in terms of the distribution of orbital states $LM_{ls}$ averaged over all atomic electron and nuclear spin quantum numbers is given by

$$\langle \ell^\prime(J)_{K_0} \rangle = \frac{1}{\ell_{1}(\ell_{J})} \frac{C_k}{2S+1} \langle \ell(J)_{K_0} \rangle . \quad (3.4.8)$$

Conversely, for only one $F$ and $J$ level being populated the orbital state distribution summed over all electron spins in terms of the coupled scheme $(LS)JM_s$ is given by

$$\langle \ell(\ell^\prime)_{K_0} \rangle = C_k \langle \ell(J^\prime J)_{K_0} \rangle . \quad (3.1.9)$$

and

$$\langle T(J)_{K_0} \rangle = \frac{1}{\ell_{2}(\ell_{J})} C_k \langle T(J^\prime)_{K_0} \rangle . \quad (3.4.10)$$

In the above equations the constant $C_k$ is

$$C_k = (-)^{I \cdot S \cdot J \cdot K} \left( \begin{array}{ccc} 2J^\prime + 1 & 2J + 1 \end{array} \right)^{\frac{1}{2}} \left\{ \begin{array}{c} I \cr J \end{array} \right\} \left\{ \begin{array}{c} I^\prime \cr J^\prime \end{array} \right\} \left\{ \begin{array}{c} K \cr S \end{array} \right\} . \quad (3.4.11)$$

If multipole moments in the hyperfine structure scheme are to be projected an additional factor

$$\sum_{FF^\prime} \left( \begin{array}{c} 2F^\prime + 1 \end{array} \right)^{\frac{1}{2}} \left( \begin{array}{c} 2F + 1 \end{array} \right)^{\frac{1}{2}} (-)^{F \cdot F^\prime \cdot J \cdot K} \left\{ \begin{array}{c} F \cr J \end{array} \right\} \left\{ \begin{array}{c} F^\prime \cr J^\prime \end{array} \right\} \left\{ \begin{array}{c} K \cr \ell \end{array} \right\} . \quad (3.4.12)$$

must be inserted.

### 3.5 Time Development and the Effect of Hyperfine Structure

The aim of electron-photon coincidence studies is to acquire the parameters which characterise the excited state at a time $t = 0$ immediately after the excitation. However, due to apparatus and state lifetime limitations, the coincidence signal is collected over a finite period of time and hence the time dependence of the coincidence signal, due
to the time evolution of the excited atom, must be taken into account. Fine structure, hyperfine structure, and time-averaging are the time development effects which must be considered.

If the lifetime of the excited state is sufficiently long for it to be neglected, an individual state $| J M_f \rangle$ with energy $E_l$ develops according to the time-evolution operator

$$ U(t) = \exp \left[ -i E_l t / \hbar \right] \quad .$$

(3.5.1)

Fano and Macek (1973) showed that, because of conservation of total spin, these time developments cannot change the rank $K$ and component $Q$ of the initial multipole moments ($t = 0$) and hence the time-evolution is simply described by

$$< T (l, t) \rangle_{KQ} = G_k(t) < T (l, 0) \rangle_{KQ}$$

(3.5.2)

if $< T (l, 0) \rangle_{KQ}$ describes the state at $t = 0$.

The $G_k(t)$ factors are **perturbation coefficients** given by

$$ G_k(t) = \sum_{J} \frac{(2J'+1)(2J+1)}{2S+1} \left[ \frac{J' J K}{S} \right]^2 \cos \left[ \frac{(E_{J'} - E_J) t}{\hbar} \right]$$

(3.5.3)

if hyperfine splitting can be neglected. Hyperfine splitting is generally neglected if $\omega_{HFS} = \Delta E_{HFS} / \hbar \ll \gamma$ where $\gamma$ is the natural line width $\gamma = \tau^{-1}$, where $\tau$ is the spontaneous lifetime of the excited state. Clearly the real multipole moments will vary periodically with time as the $G_k(t)$ do. This is a consequence of angular momentum coupling. During the collision the orbital system obtained a certain polarisation while the spin system remained unpolarised, but because of spin-orbit coupling there is a transfer of polarisation between the orbital and spin systems. Spins become polarised while the orbital system loses polarisation and hence $< T (l, t) \rangle_{KQ}$ decreases until it reaches a minimum when the spins all have maximum possible polarisation. The
process then proceeds in reverse until the initial situation is achieved at which point the cycle repeats. Clearly, the larger the (unobserved) spin, the smaller \( G_k(t) \) and hence the smaller the real multipole moments.

If hyperfine splitting must be taken into account \( (\omega_{HFS} \gg \gamma) \) the state distribution must be projected onto the (Jl)F coupled scheme and \( G_k(t) \) becomes

\[
G_k(t) = \sum_{J} \frac{\langle 2J' + 1 \rangle \langle 2J + 1 \rangle \langle 2F' + 1 \rangle \langle 2F + 1 \rangle}{\langle 2S + 1 \rangle \langle 2I + 1 \rangle} \times \left\{ F' \quad F \quad K \right\}^2 J \quad J' \quad JF(1) \text{ } (3.5.4)
\]

where \( I \) is the nuclear spin, \( F \) is the total angular momentum, and

\[
h_{J'F\cdot JF}(t) = \cos((E_{J'F} - E_{JF})t/\hbar) \text{ } (3.5.5)
\]

Clearly the time-evolution of the multipole moments now depends on the energy of the hyperfine levels, \( E_{JF} \).

In the time average all \( J' \neq J, F \) drops out of equations (3.5.3) and (3.5.4) so that equation (3.5.4) becomes a simple sum over \( J \) and \( F \) and \( h_{J'F\cdot JF} \) has to be replaced by 1. For the case that the lifetime of the excited state is very much longer than the hyperfine interaction time and fine structure is resolved (or for \( \omega_{HFS} \gg \gamma \) and \( \omega_{FS} \ll \gamma \)) then

\[
G_k = \sum_{J} \frac{(2F + 1)}{(2I + 1)} \left\{ F \quad F \quad K \right\}^2 \text{ } J \quad J \quad JF, \quad C_0 = 1 \text{ } (3.5.6)
\]

In general, though, the excited state does decay and \( \omega_{HFS}^{-1} \) is somewhere near the size of \( \gamma \), meaning that a factor of \( e^{-\gamma t} \) must be multiplied into the time dependence:

\[
h_{J'F\cdot JF}(t) = e^{-\gamma t} \cos((E_{J'F} - E_{JF})t/\hbar) \text{ } (3.5.7)
\]
Since most experiments average over time from zero to infinity, the time average values given by the time integral of $\gamma h(t)$:

$$\gamma \int_0^\infty dt e^{-\gamma t} \cos \omega t = \frac{\int_0^\infty dt e^{-\gamma t} \cos \omega t}{\int_0^\infty dt e^{-\gamma t}} = \frac{\gamma^2}{\gamma^2 + \omega^2}.$$

(3.5.8)

must be used to give

$$G_k = \sum_{J,J',F} \frac{(2J'+1)(2J+1)(2F'+1)(2F+1)}{(2S+1)(2I+1)} \frac{\langle J' \ J \ | \ F \ F' \ K \rangle^2}{\langle J' \ J \ | \ F \ F' \ S \rangle^2} \frac{\gamma^2}{\gamma^2 + \omega_{J',J}^2}.$$

(3.5.9)

### 3.6 The Stokes Parameters and the Multipole Moments

For $L \to L_2$ transitions (or $J \to J_0$ if $J$ is the relevant quantum number, as for the heavy rare gases) the photon intensity is due to decay governed by the dipole transition operator $D = \vec{e}^* \cdot \vec{r}$, where $\vec{e}$ is the polarisation vector of light emitted in the direction $\vec{k}$. The photon intensity is given by

$$I = Tr[\rho \sigma]$$

(3.6.1)

where $\sigma = D^* D$ is a detector matrix with elements

$$\sigma_{LM,LM'} = \sum_{M_0} \langle LM | \langle \vec{e}^* \cdot \vec{r} \rangle^* | L_0 M_0 \rangle \langle L_0 M_0 | \vec{e}^* \cdot \vec{r} | LM' \rangle$$

(3.6.2)

and the summation is over all final states $| L_0 M_0 \rangle$. The trace of Eq. (3.6.1) has to be carried out over all quantum numbers. The above equations are written as if no fine or hyperfine structure existed. This is not a problem since in Section 3.5 it was shown how the density matrix $\rho(L)$ is obtained from the nascent distribution.
Eq. (3.6.1) can be expressed in irreducible form in terms of its state multipole moments \( \langle l(l, l) \rangle_{KQ}^{\text{photon}} \), which are now denoted \( S_{KQ} \), by using a spherical basis for the polarisation vector:

\[
\vec{\tau} = \sum_{q=-l}^{l} \alpha_q \hat{\tau}_q
\]  

(3.6.3)

where the \( \hat{\tau}_q \) are spherical unit vectors, to obtain

\[
l \sim \sum_{KQ} \langle l(l)_{KQ} \rangle S_{KQ}^*
\]  

(3.6.4)

where multipole moments up to rank 2 can be detected. Converting to the more convenient real multipole moments and denoting the atomic real multipole moments as \( T_{KQ} \) and the real multipole moments of the detected photons as \( S_{KQ} \), the detected intensity is

\[
l = \frac{C}{3} \sum_{KQ} g_K T_{KQ} S_{KQ}^*
\]  

(3.6.5)

where

\[
g_K = (-1)^K \frac{\lambda^{(K)}(L)}{\lambda^{(0)}(L)} \frac{\chi^{(1)}}{\chi^{(1)}} \frac{\{K, L, L\}}{\{L_0, 1, 1\}} \frac{\{K, L, L\}}{\{L_0, 1, 1\}}
\]  

(3.6.6)

\[g_{0} = 1, \quad K = 0, 1, 2.\]

The constant \( C/3 \) absorbs all factors such as normalisation constants, detector efficiency, etc. For \( L \rightarrow L_0 \) or \( J \rightarrow J_0 \) transitions which are \( 1 \rightarrow 0 \) the \( g_x \) are \( g_0 = 1, g_1 = 3/2 \) and \( g_2 = 1/2 \).
Choosing the z-axis parallel to the direction of photon propagation allows the detected photon to be described by its Stokes parameters \( P_1^D, P_2^D \) and \( P_3^D \). Its density matrix is then
\[
\rho_{\text{photon}} = \langle t_q t_q^* \rangle = \frac{1}{2} \begin{pmatrix}
1 - P_3^D & 0 & -P_1^D + i P_2^D \\
0 & 0 & 0 \\
-P_1^D - i P_2^D & 0 & 1 + P_3^D
\end{pmatrix}.
\]  \( \text{(3.6.7)} \)

Comparing this equation to Table 1, only using \( S_{10}^* \) instead of \( \Xi_0 \), gives
\[
S_{00}^* = 1, \quad S_{10}^* = -P_3^D, \quad S_{20}^* = 1.
\]
\[
S_{22}^* = -\sqrt{3} P_1^D, \quad S_{22}^* = -\sqrt{3} P_2^D, \quad \text{all others are zero.} \quad \text{(3.6.8)}
\]

Alternatively, if linearly polarised light is detected it is convenient to choose the photon propagation vector \( \vec{k}_{\text{photon}} \) perpendicular to the z-axis and the z-axis parallel to the detected linear polarisation vector \( \vec{e}_{\text{linear}} \). The photon density matrix is then
\[
\rho_{\text{photon}} = \langle \alpha_q \alpha_q^* \rangle = \begin{pmatrix}
0 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 0
\end{pmatrix}
\]  \( \text{(3.6.9)} \)

and \( S_{00}^* = 1, S_{20}^* = -2 \) (all others are zero).

To account for depolarisation due to fine and hyperfine interaction and time-averaging, \( \Xi_0 \) is multiplied by the depolarisation factors \( G_x(t) \) to obtain
\[
l = \frac{1}{3} C \{ G_0 + g_2 G_2 | T_{20} - \sqrt{3} P_1^D T_{22} - \sqrt{3} P_2^D T_{22} | - g_1 G_1 P_3^D T_{10} \}
\]  \( \text{(3.6.10)} \)

for \( \vec{k}_{\text{photon}} \) parallel to \( \vec{z} \), and
\[
l = \frac{1}{3} C \{ G_0 - 2 g_2 G_2 T_{20} \}
\]  \( \text{(3.6.11)} \)

for \( \vec{k}_{\text{photon}} \) perpendicular to \( \vec{z} \), which is parallel to \( \vec{e}_{\text{linear}} \), with \( G_0 = 1 \).

If the photon frame coincides with the natural frame then Eq. (3.6.11) gives the intensities detected in the z-direction and the Stokes parameters \( (P_i) \) for the atomic
radiation can be expressed in terms of the detector Stokes parameters. The total intensity in the z-direction is

$$ I_z = I(P^D_i = +1) + I(P^D_i = -1) $$  \hspace{1cm} (3.6.12) 

and then

$$ I_z P_i = I(P^D_i = +1) - I(P^D_i = -1) $$  \hspace{1cm} (3.6.13) 

From these equations the relation of the atomic Stokes parameters to the atomic real multipole moments are found to be, in units of normalisation constant C,

$$ I_z P_1 = -(2/\sqrt{3}) g_2 C_2 T_{22} $$  \hspace{1cm} (3.6.14) 

$$ I_z P_2 = -(2/\sqrt{3}) g_2 C_2 T_{22} $$  \hspace{1cm} (3.6.15) 

$$ I_z P_3 = -(2/3) g_1 C_1 T_{10} $$  \hspace{1cm} (3.6.16) 

$$ I_z = (2/3)(C_0 + g_2 C_2 T_{20}) $$  \hspace{1cm} (3.6.17) 

The intensity emitted in the x-y scattering plane (natural frame) with polarisation parallel to the scattering plane (i.e. in the x-direction) is found by setting $P^D_i = 1$ (with $C = 1$):

$$ I^p = \frac{1}{3}(C_0 + g_2 C_2 T_{20} - \sqrt{3} g_2 C_2 T_{22}) = \frac{1}{2} I_z (1 + P_1) $$  \hspace{1cm} (3.6.18) 

The intensity in the scattering plane with polarisation perpendicular to the scattering plane (i.e. an oscillator in the z-direction) is given by Eq. (3.6.11) with $C = 1$. Since

$$ P_4 = \frac{I_{||}^p - I_{\perp}^p}{I_{||}^p + I_{\perp}^p} $$  \hspace{1cm} (3.6.19) 

it follows that

$$ g_2 C_2 T_{20} = \frac{2 P_4 - P_1 + P_1 P_4}{1 - (1 - P_1)(1 - P_4)} $$  \hspace{1cm} (3.6.20) 

and

33
\[ I_z = \frac{2(1 + P_1)}{1 - (1 - P_1)(1 - P_1)}. \]  \hspace{1cm} (3.6.21)

### 3.7 Specialisation to P→S Transitions

For general P→S transitions L = 1, L_0 = 0, g_1 = 3/2 and g_2 = 1/2 and it follows that

\[ I_z = \frac{1}{3}(2G_0 + G_2) - G_2 \rho_{00}, \]  \hspace{1cm} (3.7.1)

\[ I_z P_1^\prime = -2G_2 \text{Re}(\rho_{11}) = G_2(1 - \rho_{00})P_1, \]  \hspace{1cm} (3.7.2)

\[ I_z P_2^\prime = +2G_2 \text{Im}(\rho_{11}) = G_2(1 - \rho_{00})P_2, \]  \hspace{1cm} (3.7.3)

\[ I_z P_3^\prime = -G_1(\rho_{11} - \rho_{11}^*) = G_1(1 - \rho_{00})P_3, \]  \hspace{1cm} (3.7.4)

\[ \rho_{00} = \frac{1}{3} - \frac{2}{3G_2} \frac{2P_4^\prime - P_1^\prime + P_1^\prime P_1^\prime}{1 - (1 - P_1^\prime)(1 - P_1^\prime)}. \]  \hspace{1cm} (3.7.5)

where \( P_i \) are the reduced Stokes parameters describing the nascent charge cloud and \( P_i^\prime \) are the measured Stokes parameters. Inserting Eq. (3.6.21) and rearranging Eq. (3.7.2)-(3.7.3) gives

\[ P_i = P_i \left[ \frac{2 + G_2(1 - 3\rho_{00})}{3G_2(1 - \rho_{00})} \right], \quad i = 1, 2 \]  \hspace{1cm} (3.7.6)

and

\[ P_3 = P_3 \left[ \frac{2 + G_2(1 - 3\rho_{00})}{3G_1(1 - \rho_{00})} \right]. \]  \hspace{1cm} (3.7.7)
The absence of depolarisation (as in the Ne and Ar cases) is indicated by \( G_1 = G_2 = 1 \), which results in

\[
I_z = 1 - \rho_{00}
\]  
(3.7.8)

and illustrates that the intensity emitted in the z-direction is reduced by the relative probability of having an oscillator in the z-direction.

The density matrix can be decomposed, in the natural frame, into a part having positive and a part having negative reflection symmetry with respect to the scattering plane,

\[
\begin{pmatrix}
\rho_{11} & 0 & \rho_{1-1} \\
0 & \rho_{00} & 0 \\
\rho_{-11} & 0 & \rho_{-1-1}
\end{pmatrix} = (1 - \rho_{00}) \frac{1}{2} \begin{pmatrix}
1 - \rho_{33} & 0 & -\rho_1 + i\rho_2 \\
0 & 0 & 0 \\
-\rho_1 - i\rho_2 & 0 & 1 + \rho_{33}
\end{pmatrix} \\
+ \rho_{00} \begin{pmatrix}
0 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 0
\end{pmatrix}
\]  
(3.7.9)

and can be expressed in terms of the excitation amplitudes \( \alpha_{m_{s_2}M_{J}m_i} \) (where \( m_{s_1} \) and \( m_{s_2} \) are the initial and final spin projection quantum numbers) as

\[
\rho_{M_J M_{J'}} = \sum_{m_{s_1} m_{s_2}} \alpha_{m_{s_2} M_{J} m_{s_1}} \alpha_{m_{s_2} M_{J'} m_{s_1}}^*.
\]  
(3.7.10)

The excitation amplitudes are normalised to the total differential cross section

\[
\sigma(\theta) = \frac{k_{\text{out}}}{k_{\text{in}}} \sum_{M_{J} M_{J'}} |f_{M_{J} M_{J'} m_{s_1}}|^2
\]  
(3.7.11)

through

\[
\alpha_{m_{s_2} M_{J} m_{s_1}} = f_{M_{J} M_{J'} m_{s_1}} \left( \frac{k_{\text{out}}}{k_{\epsilon}} \right)^{\frac{1}{2}} \sigma(\theta)^{-\frac{1}{2}}
\]  
(3.7.12)

with normalisation

\[
1 = \sum_{M_{J} M_{J'} m_{s_1}} |\alpha_{m_{s_2} M_{J} m_{s_1}}|^2.
\]  
(3.7.13)
Symmetry demands that
\[ \alpha_{\frac{1}{2}0^{-\frac{1}{2}}} = \alpha_{\frac{1}{2}0^{\frac{1}{2}}} \]  \hspace{1cm} (3.7.11)
and the non-zero amplitudes are then
\[ \alpha_{\frac{1}{2}0^{-\frac{1}{2}}}, \alpha_{\frac{1}{2}1^{-\frac{1}{2}}} \text{ and } \alpha_{\frac{1}{2}1^{\frac{1}{2}}} \, . \]

The density matrix elements are then
\[ \rho_{00} = 2 \beta_{\frac{1}{2}0^{-\frac{1}{2}}}^2, \quad M_j = 0 \]  \hspace{1cm} (3.7.15)
and
\[ \rho_{M_jM_{-j}} = \alpha_{\frac{1}{2}M_jM_{-j}}^2 + \alpha_{-\frac{1}{2}M_jM_{-j}}^2, \quad M_j, M_{-j} = \pm \frac{1}{2} \, . \]  \hspace{1cm} (3.7.16)

For the fully coherent case a collisionally excited $^1P_1$ state may be described as a superposition of states. Since, in this case, no spin flip can occur the final state must have the same reflection symmetry as the initial, positive reflection symmetry, $^3S_1$ state. Consequently, only p-orbitals oscillating in the scattering plane can be excited and the $^1P_1$ state may be written as
\[ ^1P_1 = \alpha_+ |\sigma^{-} \rangle + \alpha_- |\pi^{-} \rangle = \alpha_1 |1^{-} \rangle + \alpha_{-1} |1^{+} \rangle \]  \hspace{1cm} (3.7.17)
where the $M_j$ basis is in the natural frame. The excitation amplitudes are related by
\[ \alpha_+ = \frac{1}{\sqrt{2}} (\alpha_1 - \alpha_{-1}) \]  \hspace{1cm} (3.7.18)
\[ \alpha_- = \frac{i}{\sqrt{2}} (\alpha_1 + \alpha_{-1}) \]  \hspace{1cm} (3.7.19)
\[ \alpha_{+1} = \frac{1}{\sqrt{2}} (\alpha_+ - i\alpha_-) \]  \hspace{1cm} (3.7.20)
Clearly the notation for the fully coherent case is much more compact than for the previous general P→S case, and hence the fully coherent case is used to illustrate the relations of the Stokes parameters to the frame independent parameters \( \gamma \), \( \rho' \), \( l' \), and \( \rho_{\infty} \).

Since the atomic state can be represented as a superposition of two circular states of angular momentum +1 and -1 (\( \hbar = 1 \)), the expectation value of the angular momentum perpendicular to the scattering plane is

\[
|l_\perp|^2 = <l_z> = |\alpha_1|^2 - |\alpha_{-1}|^2 .
\] (3.7.21)

The circular polarisation observed along the z-axis originates from the two circular states emitting left-handed (\( \alpha_1 \)) and right-handed (\( \alpha_{-1} \)) circularly polarised light and is given by

\[
P_3 = \frac{l_R - l_L}{l_R + l_L} = |\alpha_{-1}|^2 - |\alpha_1|^2 = -|l_\perp|^2 .
\] (3.7.22)

Assuming the charge cloud is composed of two linear oscillators with amplitudes \( \alpha_0 \) and \( \alpha_{n^*} \), the Stokes parameters for the linear polarised light emitted perpendicular to the scattering plane are

\[
P_1 = \frac{l(0^\circ) - l(90^\circ)}{l(0^\circ) + l(90^\circ)} = |\alpha_0|^2 - |\alpha_{n^*}|^2 = -2\text{Re}(\alpha_1\alpha_{-1}^*)
\] (3.7.23)

and

\[
P_2 = \frac{l(45^\circ) - l(135^\circ)}{l(45^\circ) + l(135^\circ)} = 2\text{Re}(\alpha_0\alpha_{n^*}^*) = 2\text{Im}(\alpha_1\alpha_{-1}^*)
\] (3.7.24)

Andersen et al (1988) related \( \gamma \) to \( P_1 \) and \( P_2 \) by using

\[
\psi \sim \alpha_0 \cos \phi + \alpha_{n^*} \sin \phi
\] (3.7.25)
as the wavefunction of the charge cloud in some direction \( \phi \), with respect to the x-axis, in the scattering plane. The angular part of the electron density is then
\[ \psi^2 = \frac{1}{2} - \left[ \cos 2\phi \Re(\alpha_1 \alpha_{-1}^*) - \sin 2\phi \Im(\alpha_1 \alpha_{-1}^*) \right] \]

\[ = \frac{1}{2} + \alpha_1 \cdot \alpha_{-1} \cdot \cos 2(\phi - \gamma) \quad (3.7.26) \]

where

\[ \alpha_1 \alpha_{-1}^* = -\alpha_1 \cdot \alpha_{-1} \cdot e^{i \gamma} \quad (3.7.27) \]

Clearly the maximum charge cloud density is in the direction \( \phi = \gamma \) and thus the charge cloud alignment angle is linearly related to the phase difference between \( \alpha_1 \) and \( \alpha_{-1} \):

\[ \gamma = -\frac{1}{2} \arg(\alpha_1 \alpha_{-1}^*) \quad (3.7.28) \]

Andersen et al. (1988) introduced the linear polarisation \( P_i \) as the relative difference between the length and width of the charge cloud, or maximum and minimum densities \( \psi_{\text{max}}^2 \) and \( \psi_{\text{min}}^2 \):

\[ P_i = \frac{\psi_{\text{max}}^2 - \psi_{\text{min}}^2}{\psi_{\text{max}}^2 + \psi_{\text{min}}^2} = 2 |\alpha_1| |\alpha_{-1}| = (P_1^2 + P_2^2)^{\frac{1}{2}} \quad (3.7.29) \]

A little algebra results in

\[ P_1 + iP_2 = P e^{2i\gamma} \quad (3.7.30) \]

and

\[ \gamma = \frac{1}{2} \tan^{-1}\left(\frac{P_2}{P_1}\right) \quad (3.7.31) \]

Writing the Stokes parameters in terms of amplitudes and applying the amplitude normalisation yields

\[ P_1^2 + P_2^2 + P_3^2 = P_1^2 + l_+^2 = 1 \quad (3.7.32) \]

Finally, Andersen et al. (1988) write the charge cloud distribution as
\[ \psi^2 = 1 + P_i \cos[2(\phi - \gamma)] \quad . \quad (3.7.33) \]

The density matrix for the fully coherent case is

\[ \rho_{mn} = \alpha_m \alpha_n^{*} = \frac{1}{2} \begin{pmatrix} 1 - P_3 & 0 & -P_1 + iP_2 \\ 0 & 0 & 0 \\ -P_1 - iP_2 & 0 & 1 + P_3 \end{pmatrix} . \quad (3.7.34) \]

This is the result obtained if \( \rho_{00} = 0 \) is used in the density matrix for a general P \( \rightarrow \) S transition (Eq.(3.7.9)).

In fact, the non-coherent case exactly parallels the coherent case and the charge cloud distribution can be written (Andersen et al (1988)) in spherical coordinates as

\[ Y(0, \phi) = (1 - \rho_{00})^{\frac{1}{2}} (1 + P_i \cos[2(\phi - \gamma)]) \sin^2 0 + \rho_{00} \cos^2 0 \quad . \quad (3.7.35) \]

Comparison with Eq. (3.7.33) indicates that \( \rho_{00} \) may be considered a height parameter for the charge cloud. In the y-direction (in the scattering plane) \( P_4 \leq 1 \) and

\[ \rho_{00} = \frac{(1 + P_1)(1 - P_4)}{1 - (1 - P_1)(1 - P_4)} . \quad (3.7.36) \]

For the incoherent case the parts with positive reflection symmetry must be separated from those with negative symmetry. Hence a superscript "+" is used to indicate positive reflection symmetry. Then

\[ P_i^+ = (P_1^2 + P_2^2)^{\frac{1}{2}} . \quad (3.7.37) \]

Writing \( l_\perp = -P_3 \) the angular momentum perpendicular to the scattering plane is

\[ L_\perp = -P_3(1 - \rho_{00}) = L^+_i(1 - \rho_{00}) . \quad (3.7.38) \]

The degree of polarisation (or coherence) is

\[ P^* = (P_1^2 + P_2^2 + P_3^2)^{\frac{1}{2}} \leq 1 \quad (3.7.39) \]

with equality holding only for the fully coherent case.
Finally, the length \( l \), width \( w \) and height \( h \) of the charge cloud are given by

\[
l = \frac{1}{2} (1 - \rho_{oo})(1 + p_1^*) \quad (3.7.10)
\]

\[
u = \frac{1}{2} (1 - \rho_{oo})(1 - p_1^*) \quad (3.7.11)
\]

and

\[
h = \rho_{oo} \quad (3.7.12)
\]

with

\[
p_1^* = \frac{l - u}{l + u} \quad (3.7.13)
\]

and

\[
l + u + h = 1 \quad (3.7.14)
\]
Chapter IV
THEORETICAL VOLUME EFFECT MODELS

4.1 Introduction

The general theory of electron-photon coincidence experiments takes no account of the fact that in a real experiment the interaction region formed by the intersection of the electron beam and the atomic gas beam has finite volume. Figure 6 illustrates how a scattering event in this finite volume may correspond to a coordinate frame slightly different than the natural frame used in the theoretical formulation. In this geometry, for any scattering event, the x-axis is chosen coincident with the axis of the electron beam and the y-axis is chosen such that the electron detector entrance aperture is in the x-y plane (the scattering plane). In Figure 6 a scattering event is considered to have occurred at the "top" of the electron beam at a point vertically displaced from the nominal scattering plane. In this case the scattered electron passes from the top of the electron beam through the entrance aperture and defines the $x' - y'$ plane as the scattering plane.

Clearly the primed scattering plane is rotated with respect to the nominal scattering plane and the scattering angle is slightly different from the nominal scattering angle. Hence the intensity and polarisation of radiation from the event at the top of the electron beam will be somewhat different from that from the center of the interaction region, and could affect measured quantities. It is also possible that the electron beam is displaced by a small amount due to mechanical misalignment of the system or by electrostatic deflection and so in order to develop models describing the effects of the finite volume of the interaction region the nominal scattering plane is defined by the axis of the electrostatic lens elements of the electron gun (x-axis) and the position of the center of the entrance aperture of the electron detection system.
Figure 6. Schematic diagram corresponding to analytical model. Electrons scattered from the interaction region into the electron detection system from the center or top of the electron beam define the $x - y$ and $x' - y'$ scattering planes, respectively. The planes are rotated from one another through an angle $\beta$.

In addition to possible effects due to the size and displacement of the electron beam other finite collision volume effects may be due to the finite size of the gas beam, the preferential acceptance of scattered electrons from away from the nominal scattering center due to misfocussing of the electron detection system, and the variation of intensity and polarisation of the emitted radiation as a function of scattering angle within the interaction volume. Other possible effects could originate on the detection
side and may be due to the finite acceptance angle of the photon detectors and the finite acceptance angle of the electron detection system (the diameter of the entrance aperture).

In this chapter two different models are developed for the purpose of quantitatively studying the effects of a finite interaction region: (1) an approximate analytical model based on the assumption that only the finite size of the electron beam is important, and (2) a numerical model which incorporates all the previously mentioned possible effects. To develop these models the C-matrix formalism of Nienhuis (1980) is employed and hence it warrants a section proceeding the model development.

4.2 The C-Matrix Formalism

In their groundbreaking theoretical treatment of the electron-photon coincidence experiment Macek and Jaecks (1971) determined that for coincident scattering of an electron into solid angle element $d\Omega_e$ and emission of a photon of polarisation $\mathbf{\hat{e}}$ into element $d\Omega_p$ the coincidence rate per unit scattering volume is

$$N(\mathbf{\hat{e}}) = \frac{\omega^3}{2\pi \hbar c^3} \ d\Omega_e \ d\Omega_p \ Tr \left( \mathbf{\hat{e}}^\dagger \cdot \mu_{ef} \rho_e \mu_{if} \cdot \mathbf{\hat{e}} \right) \quad (4.2.1)$$

where $\omega$ is the frequency of the emitted photons, $\mu_{if}$ is the electric dipole operator between the initial excited state $|i\rangle$ and the final state $|f\rangle$ of the atom, $\rho_e$ is the density matrix of the excited atoms decaying within the resolution time of the coincidence apparatus, $T$ is the data accumulation time interval, and $r_f$ is the branching ratio of the final state. Nienhuis (1980) introduced the 3 x 3 Cartesian matrix $\mathbf{C}$ such that the coincidence rate per unit volume is given by

$$N(\mathbf{\hat{e}}) = \mathbf{\hat{e}}^\dagger \cdot \mathbf{C} \cdot \mathbf{\hat{e}} \quad (4.2.2)$$

The matrix $\mathbf{C}$ determines the intensity and polarisation of photons emitted in any direction, and is therefore particularly suited for studying finite volume effects.
Because the C-matrix is related to the density matrix of the excited atoms, any anisotropy of the excited state density matrix will result in a similar anisotropy of the intensity and polarisation of the emitted radiation. The exact relation between \( \rho \) and \( \mathcal{C} \) can be found by expanding both \( \rho \) and \( \mathcal{C} \) in irreducible spherical tensors, and is not presented here. A general expression for \( \mathcal{C} \) can be found by applying hermiticity and reflection symmetry of \( \rho \), and Nienhuis (1980) has shown that, in the natural frame, \( \mathcal{C} \) is given by

\[
\mathcal{C} = \frac{I_z}{2} \begin{pmatrix}
1 + P_1 & P_2 + i P_3 & 0 \\
0 & 1 - P_1 & 0 \\
0 & 0 & C_{33}
\end{pmatrix}
\]

(1.2.3)

where

\[
C_{33} = \frac{(1 + P_1)(1 - P_3)}{(1 + P_1)}
\]

(1.2.4)

and \( I_z \) is the coincidence rate for the radiation in the z-direction, \( P_1, P_2 \) and \( P_3 \) are the Stokes parameters of the radiation emitted in the z-direction, and \( P_3 \) is the linear polarisation of the radiation emitted in the +\( \gamma \) (or -\( \gamma \)) direction. This form of the C-matrix holds for any atomic dipole transition assuming that the incident electrons are unpolarised and the atoms in the initial state are unpolarised and that the spin of the scattered electrons is not observed.

The common 2 x 2 polarisation matrix of the radiation emitted in a particular direction is found from the matrix elements of \( \mathcal{C} \) between polarisation vectors orthogonal to this direction. For example, the \( x - \gamma \) submatrix of \( \mathcal{C} \) gives the 2 x 2 polarisation matrix of the radiation in the z-direction. In Eq. (4.2.1) an ideal situation is assumed in the sense that the photon detector is sensitive to light with a particular polarisation \( \hat{e} \) only, so it is convenient to generalise this equation for a detector with arbitrary polarisation sensitivity. Using a right-handed orthogonal set of unit vectors
\{ \hat{e}_1, \hat{e}_2, \hat{e}_3, \hat{e}_4 \} \) the 2 x 2 density matrix for the photons emitted in the direction 
\( \vec{n} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) \) has elements
\[
\epsilon_{ij} = \hat{e}_i \cdot \hat{e}_j .
\] (4.2.5)

This density matrix is given with respect to the linear polarisation vectors \( \hat{e}_1 \) and \( \hat{e}_2 \) and
is known as the coherency matrix (O'Neill 1963). In terms of the Stokes parameters of
the light emitted in the direction \( \vec{n} \) the coherency matrix is
\[
\boldsymbol{C} = \frac{1}{2} \begin{pmatrix}
1 + P_1(\vec{n}) & P_2(\vec{n}) + iP_3(\vec{n}) \\
P_2(\vec{n}) - iP_3(\vec{n}) & 1 - P_1(\vec{n})
\end{pmatrix} .
\] (4.2.6)

According to coherency matrix formalism the transformation law for the density
(coherency) matrix for light passing through an optical component is (O'Neill 1963)
\[
\boldsymbol{C}' = \mathcal{A}_c \mathcal{A}_c^* .
\] (4.2.7)

where \( \mathcal{A} \) is the Jones matrix representing the operation of the optical component.
Introducing a 2 x 2 efficiency matrix \( \mathcal{E} \) for the photon detector behind the optical
component the detected signal follows from
\[
I = Tr(\mathcal{E} \mathcal{C}') = Tr(\mathcal{E} \mathcal{A}_c \mathcal{A}_c^*) .
\] (4.2.8)

In most optical systems used in coherence experiments (see Slevin and Chwirot
1990) the Stokes parameters are extracted from measurement of the detected signal
as a function of the angle of one or more rotatable optical components. In this work
single and double-reflection polarisation analysers (Westerveld et al 1985) that employ
reflection from gold coated mirrors are used.

4.3 Approximate Analytical Model

In this section analytical expressions for the measured Stokes parameters
\( (P_i^M, i = 1, 2, 3, 4) \) are derived. This model is based on the assumption that only
the finite size of the electron beam is important in regard to the effects of a finite
interaction volume and neglects the variation of the Stokes parameters over the small

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range of scattering angles within the interaction region. Although this is not always justified it has been found that this model compares well with the more sophisticated numerical model for a number of situations and the analytical formulae provide some insight into the effect of a finite interaction region. In addition, as discussed later, it is possible to invert the expressions obtained for \( P_{i}^{M} \) in order to obtain actual values of \( P_{i} \). So far this has not proved to be possible with any model and certainly not with a more sophisticated one.

Consider, again, Figure 6. For electrons scattered from a point with coordinates \((0, y_{p}, z_{p})\) off the axis of the electron beam a collision plane is defined by the \( x \) and \( y \) axes (the particular case shown in the figure is for electrons scattered from the top of the electron beam). For this point the polarisation matrix \( C' \) with respect to the natural \((x', y', z')\) coordinate frame is

\[
C' = \frac{i}{2} \begin{pmatrix}
1 + P_{1}' & P_{2}' + iP_{3}' & 0 \\
0 & 1 - P_{1}' & P_{2}' - iP_{3}' \\
0 & 0 & C_{33}'
\end{pmatrix}
\]  

(4.3.1)

As the scattered electrons pass through the aperture into the electron detection system, at a distance \( R_{e} \) from the scattering center, the \((x', y', z')\) frame is rotated with respect to the \((x, y, z)\) frame over an angle \( \beta \) given by

\[
\tan \beta = \frac{z_{p}}{R - y_{p}} = \frac{z_{p}}{R_{e} \sin \theta - y_{p}}
\]

(4.3.2)

where \( \theta \) is the electron scattering angle. The polarisation matrix with respect to the \((x, y, z)\) frame can then be found using the rotation matrix for a rotation over \( \beta \) about the x-axis:

\[
R(\beta) = \begin{pmatrix}
1 & 0 & 0 \\
0 & \cos \beta & -\sin \beta \\
0 & \sin \beta & \cos \beta
\end{pmatrix}
\]

(4.3.3)

so that:
\[ C'(\beta) = R'(\beta) \overline{C R(\beta)} \quad (4.3.1) \]

or

\[
\overline{C} = \frac{I_2}{2} \begin{pmatrix}
1 - P_3 & (P_2 - iP_3) \cos \beta & -(P_2 - iP_3) \sin \beta \\
(P_2 - iP_3) \cos \beta & (1 - P_1) \cos^2 \beta - C_{33} \sin^2 \beta & -(1 - P_1) \sin \beta \cos \beta - C_{33} \sin \beta \cos \beta \\
-(P_2 - iP_3) \sin \beta & -(1 - P_1) \sin \beta \cos \beta + C_{33} \sin \beta \cos \beta & (1 - P_1) \sin^2 \beta + C_{33} \cos^2 \beta
\end{pmatrix}
\quad (4.3.5)
\]

Assuming that radiation from individual atoms at various points in the interaction region adds incoherently, the intensity and polarisation of the emitted radiation from all excited atoms in the interaction region can be derived by integrating (or averaging) \( C'(\beta) \) over the interaction region. This approach is justified because the distance to the photon detectors is much larger than the size of the interaction region. Making the assumption that variation of scattering angle for points within the interaction region is negligible, the averaging may be restricted to points located within the electron beam radius \( r \). Integration yields \( \overline{\sin \beta} = 0, \overline{\sin \beta \cos \beta} = 0 \), and

\[
\overline{C} = \frac{I_2}{2} \begin{pmatrix}
1 + P_1 & (P_2 + iP_3) \nu & 0 \\
(P_2 - iP_3) \nu & (1 - P_1)(1 - \mu) + C_{33} \mu & 0 \\
0 & 0 & (1 - P_1) \mu + C_{33}(1 - \mu)
\end{pmatrix}
\quad (4.3.6)
\]

where

\[
\mu = \overline{\sin^2 \beta} = \frac{r^2}{4R^2} = \frac{r^2}{4R_s^2 \sin^2 \theta_s}
\]

\[
1 - \mu = \overline{\cos^2 \beta} = 1 - \frac{r^2}{4R^2}
\]

\[
\nu = \overline{\cos \beta} = 1 - \frac{1}{8} \frac{r^2}{R^2} - \frac{1}{64} \frac{r^4}{R^4}
\quad (4.3.7)
\]
Ignoring the opening angle of the photon detectors by assuming that in the
z-direction all detected radiation is emitted parallel to the z-axis (i.e. \( I_z = I_z \)) the
polarisation is described by the \( N - \gamma \) submatrices of \( \overline{C} \) and \( \overline{C} \):
\[
\overline{C}_z \sim \begin{pmatrix} 1 + P_1 & (P_2 + iP_3) \nu \\ (P_2 - iP_3) \nu & (1 - P_1)(1 - \mu) + C_{33} \overline{11} \end{pmatrix} \sim \begin{pmatrix} 1 + P_1 & P_2 + iP_3 \\ P_2 - iP_3 & 1 - P_1 \end{pmatrix} .
\] (4.3.8)

Normalising each of these matrices to their traces and taking sums and differences, for example \( c_{12} + c_{21} \), gives the relation of the actual Stokes parameters \( P_i \) to the measured Stokes parameters \( P_i^M \) (previously denoted \( P_i \)):
\[
P_i^M = \frac{1 - \left( \frac{1 - P_1}{1 - P_i} \right)(1 - \mu)}{1 + \left( \frac{1 - P_1}{1 - P_i} \right)(1 - \mu)} - \frac{\left( \frac{1 - P_4}{1 - P_i} \right)\mu}{1 + \left( \frac{1 - P_4}{1 - P_i} \right)\mu} \] (4.3.9)

and
\[
P_i^M = \frac{2 \nu P_i}{(1 + P_1) \left[ 1 + \left( \frac{1 - P_1}{1 - P_i} \right)(1 - \mu) + \left( \frac{1 - P_4}{1 - P_i} \right)\mu \right]} , \quad i = 2, 3 \] (4.3.10)

where \( \mu \) and \( \nu \) are as given in Eq. (4.3.7).

Analogously the measured \( P_4^M \) parameter is found from the \( N - \cdot \cdot \) submatrices of
\( \overline{C} \) and \( \overline{C} \):
\[
\overline{C}_\gamma \sim \begin{pmatrix} 1 + P_1 & 0 \\ 0 & (1 - P_1)\mu + C_{33}(1 - \mu) \end{pmatrix} \sim \begin{pmatrix} 1 + P_1 & 0 \\ 0 & C_{33} \end{pmatrix} .
\] (4.3.11)

Clearly no circularly polarised radiation is emitted in this direction and the linear
polarisation is
\[
P_4^M = \frac{1 - \left( \frac{1 - P_1}{1 - P_i} \right)\mu}{1 + \left( \frac{1 - P_4}{1 - P_i} \right)\mu} \left( \frac{1 - P_4}{1 - P_i} \right)(1 - \mu) \] (4.3.12)
In the case of helium $P_4 = 1$ and Eq. (4.3.12) reduces to

$$P^M_i = \frac{1 - \left(\frac{1-P_i^M}{1-P_i^M}\right) \mu}{1 + \left(\frac{1-P_i^M}{1-P_i^M}\right) \mu}. \quad (4.3.13)$$

Equation (4.3.12) illustrates two important points with regard to spurious effects on $P^M_i$ due to a finite interaction region. First, since the effect depends on

$$\mu = \frac{\sin^2 \beta}{r^2 / \lambda R^2 \sin^2 \theta \alpha},$$

it is only likely to be significant for small scattering angles where the variation of $\mu$ is the largest. Second, an amplifying effect occurs for $P_i < 0$, especially when $P_i$ approaches -1. For excitation of the rare gas atoms at intermediate energies $P_i$ becomes negative at small scattering angles, where the variation of $\mu$ can become significant, and where part of the measurements in this work were carried out. Thus, in a graphical picture, an effect on $P^M_i$ is expected for small scattering angles, especially if the charge cloud is very elongated and at those angles where the charge cloud is facing the $P_i$ photon detector end on.

From Eq. (4.3.9) a much smaller effect is expected on $P^M_i$ since for the rare gas atoms $P_4$ is anticipated to be close to unity over the full range of scattering angles.

Equations (4.3.9), (4.3.10) and (4.3.12) can be readily inverted to extract the actual $P$, parameters from the measured $P^M_i$ parameters:

$$P_1 = \frac{1 - \left(\frac{1-P_1^M}{1-P_1^M}\right) \left(\frac{1-\mu}{1-2\mu}\right) - \left(\frac{1-P_4^M}{1-P_4^M}\right) \left(\frac{\mu}{1-2\mu}\right)}{1 + \left(\frac{1-P_1^M}{1-P_1^M}\right) \left(\frac{1-\mu}{1-2\mu}\right) + \left(\frac{1-P_4^M}{1-P_4^M}\right) \left(\frac{\mu}{1-2\mu}\right)} \quad (4.3.14)$$

$$P_i = \frac{2 \mu P_i^M}{(1 + P_i^M) \left[(1 + P_i^M) - \mu (2 + P_i^M + P_4^M)\right] \nu}, \quad \ell = 2, 3 \quad (4.3.15)$$

$$P_4 = \frac{1 - \left(\frac{1-P_4^M}{1-P_4^M}\right) \left(\frac{\mu}{1-2\mu}\right) - \left(\frac{1-P_1^M}{1-P_1^M}\right) \left(\frac{1-\mu}{1-2\mu}\right)}{1 + \left(\frac{1-P_4^M}{1-P_4^M}\right) \left(\frac{\mu}{1-2\mu}\right) + \left(\frac{1-P_1^M}{1-P_1^M}\right) \left(\frac{1-\mu}{1-2\mu}\right)} \quad (4.3.16)$$

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where \( \mu \) and \( \nu \) are given in Eq. (4.3.7).

4.4 Numerical Model

A computer program has been developed to numerically simulate the effect of a finite interaction region on the measured Stokes parameters. In this model the finite volume of the intersection of the electron beam and the gas jet is represented by an array of discrete scattering points. For each point the intensity of the light detected by either the channeltron perpendicular to the nominal scattering plane or the channeltron in the nominal scattering plane is calculated. The total signals are obtained by summing over all the array points.

For every array point in the interaction region the calculation proceeds through the following steps:

1. Calculation of the set of unit vectors \( \{ \delta_1, \delta_2, \delta_3 \} \) for the \( \{x', y', z'\} \) coordinate frame. For a particular array point the scattering plane is defined by the direction \( \delta_1 \) of the incident electron and the direction \( \hat{n}_z \) of the scattered electron passing through the entrance aperture of the electron detection system, as illustrated in Figure 7. The vectors \( \delta_2 \) and \( \delta_3 \) are obtained by:

\[
\delta_3 = \frac{\delta_1 \times \hat{n}_z}{|\delta_1 \times \hat{n}_z|} \quad \text{and} \quad \delta_2 = \delta_3 \times \delta_1.
\]

2. Evaluation of the scattering angle \( \theta \) (the angle between \( \delta_1 \) and \( \hat{n}_z \)) and the corresponding theoretical Stokes parameters. The theoretical Stokes parameters are found by interpolation from the tabulated results from Bartschat and Madison (1987) for the noble gas atoms, and from Madison and Winters (1983) for helium.
Figure 7. Schematic diagram relevant to numerical model.

(3) Calculation of the radiation pattern in the lab frame by transformation of the \( C \) matrix given by Eq. (4.3.1) from the \( \{x', y', z'\} \) frame to the \( \{x, y, z\} \) laboratory frame. This is performed using

\[
C = R^* C' R
\]  \hspace{1cm} (4.4.2)

where the elements of the rotation matrix \( R \) are given by the components of the unit vectors \( \hat{b}_1, \hat{b}_2, \hat{b}_3 \) with respect to the unit vectors \( \hat{a}_1, \hat{a}_2, \hat{a}_3 \) corresponding to the \( \{x, y, z\} \) frame:

\[
R_{ij} = \hat{b}_i \cdot \hat{a}_j \]  \hspace{1cm} (4.4.3)
(4) Calculation of the Stokes parameters for the radiation emitted towards the photon detectors perpendicular to and in the nominal scattering plane. The photon detector in the nominal collision plane is at an angle \( \theta_1 \) with respect to the electron beam (the x-axis), the value of which can be set to 90\(^\circ\), 45\(^\circ\), or any other angle in the program. The 2 x 2 density matrix \( \mathcal{C} \), specifying the Stokes parameters for the radiation emitted in the direction \( \mathbf{n} \), can be obtained from Eq. (4.2.5).

For the photon detector in the scattering plane at an angle \( \theta_1 \) with the x-axis,

\[
\hat{\epsilon}_1 = \begin{pmatrix} \sin \theta_1 \\ \cos \theta_1 \\ 0 \end{pmatrix} \quad \text{and} \quad \hat{\epsilon}_2 = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}
\]

and the photon density matrix becomes

\[
\mathcal{C} = \frac{I_2}{2} \begin{pmatrix} 1 - P_1 \cos 2\theta_4 + P_2 \sin 2\theta_4 & 0 \\ 0 & C_{33} \end{pmatrix}.
\]

Note that this \( C_{33} \) is not necessarily the quantity defined in Eq. (4.2.4). That value corresponded to the special case of \( \theta_4 = 90^\circ \). For an arbitrary value of \( \theta_4 \), the linear polarization of the radiation emitted in the direction \( \mathbf{n} \) in the scattering plane is

\[
P_4(\theta_4) = \frac{1 - P_1 \cos 2\theta_4 + P_2 \sin 2\theta_4 - C_{33}}{1 - P_1 \cos 2\theta_4 + P_2 \sin 2\theta_4 + C_{33}}.
\]

which yields

\[
C_{33} = \frac{(1 - P_1 \cos 2\theta_4 + P_2 \sin 2\theta_4)(1 - P_4(\theta_4))}{(1 + P_4(\theta_4))}.
\]

In the special case of \( \theta_4 = 90^\circ \), Eq. (4.4.7) reduces to Eq. (4.2.4).

(5) Calculation of the intensity of radiation detected by the photon detector in the nominal scattering plane. In order to obtain the intensity from Eq. (4.2.8) the optical components in the detector must be taken into account. For a reflecting surface as
employed in the detectors in this work, the Jones matrix takes the form (Westerveld et al 1985):

$$\mathbf{J} = \begin{pmatrix} r_p \exp\{i\delta_p\} & 0 \\ 0 & r_s \exp\{i\delta_s\} \end{pmatrix}. \quad (4.1.8)$$

Here $r_p \exp\{i\delta_p\}$ and $r_s \exp\{i\delta_s\}$ are the complex reflection coefficients and $\delta_p$ and $\delta_s$ are the phase shifts for light polarised parallel to and perpendicular to the plane of reflection, respectively. For reflection from a flat surface the reflection coefficients can be expressed in terms of the complex index of refraction ($\bar{n} = n + ik$) of the surface medium. The mirrors used in this work are gold coated, and the angle of incidence is chosen such that $\cos(\delta_p - \delta_s) = 0$. Assuming that the photon detector is insensitive to polarisation and has an efficiency $e_0$ for the total intensity of the incident radiation, the efficiency matrix is $e_0 \mathbf{1}$ and the detected signal becomes

$$I(\alpha) = e_0 \sum_{i,j,k-1} A_{ki} \mathbf{\tilde{e}}_i \cdot \mathbf{\tilde{C}} \cdot \mathbf{\tilde{e}}_j A_{kj}^{\ast} \quad (4.4.9)$$

where $\alpha$ is the angle between the reflection plane and the nominal scattering plane. As discussed in Section 3.3 the linear polarisation is obtained from measurements of intensity with the mirror reflecting in the scattering plane and perpendicular to the scattering plane. To apply Eq. (4.4.9) the vectors $\mathbf{\tilde{e}}_1$ and $\mathbf{\tilde{e}}_2$ are chosen parallel to and perpendicular to the reflection plane defined by the unit vectors $\mathbf{\hat{n}}$ and $r\mathbf{n}$ (normal vector of the mirror), as displayed in Figure 7.

For reflection parallel to the reflection plane $\mathbf{\tilde{e}}_1 = \mathbf{\hat{n}}$ and $\mathbf{\tilde{e}}_2 = \mathbf{\hat{z}}$ and the intensity is given by

$$I(0^\circ) = e_0 (R_p C_{33} + R_s C_{11}) \quad (4.4.10)$$

where $R_p = (r_p)^2$ and $R_s = (r_s)^2$ are the reflection coefficients. Reflection perpendicular to the reflection plane yields the intensity
\[ I(90^\circ) = e_p (R_p C_{11} + R_c C_{13}) \]  \hspace{1cm} (1.1.11)

and thus

\[ \frac{I(0^\circ) - I(90^\circ)}{I(0^\circ) + I(90^\circ)} = \frac{R_z - R_p}{R_z + R_p} \frac{C_{11} - C_{33}}{C_{11} + C_{33}} = \eta P_4 \]  \hspace{1cm} (1.1.12)

where

\[ \eta = \frac{R_z - R_p}{R_z + R_p} \]  \hspace{1cm} (1.1.13)

is the polarisation efficiency of the mirror. The quantities \( R_z \) and \( R_p \) are functions of the angle of incidence of the radiation as well as functions of \( n \), the real part of the index of refraction, and \( k \), the extinction coefficient, both of which are dependent on the wavelength of the detected radiation as well as the mirror surface material.

The linear polarisation in the nominal scattering plane is obtained from

\[ P_i^M(0) = \frac{1}{\eta} \sum \frac{I(0^\circ) - I(90^\circ)}{I(0^\circ) + I(90^\circ)} \]  \hspace{1cm} (1.1.14)

where the summation is over the array of scattering points in the interaction region.

(6) Calculation of the intensity of the radiation detected by the photon detector perpendicular to the nominal scattering plane. A similar method to that in step (5) could be used for the signal obtained by this detector, however, the measured Stokes parameter \( P_i^M \) is obtained directly from the \( 2 \times 2 \) \( x - y \) submatrix of \( C \) in Eq. (4.4.2):

\[ P_i^M = \sum \frac{C_{11} - C_{22}}{C_{11} + C_{22}} \]  \hspace{1cm} (4.1.15)

This amounts to assuming that only radiation emitted parallel to the z-axis from an arbitrary point in the interaction region is detected. This is a legitimate approximation in view of the large distance (about 120 mm) of the detector from the interaction region. In this case there is actually a polarisation efficiency which must be included, but it is assumed that only one angle of incidence is significant and hence the value is

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the same for all array points.

In these calculations the array of discrete scattering points is generated by starting with a three-dimensional cubic lattice of array points, with the distance between array points to be set by the program user. The finite size of the electron beam is modelled by rejecting all array points outside the radius of the electron beam. Apart from the radius, the displacement position of the electron beam in the $y-z$ plane can be specified in the program. In addition a divergent (or convergent) electron beam can be modelled by deforming the lattice of array points towards a focal point of the electron beam specified by the user.

The finite size and density of the gas jet is implemented by assigning a weight factor to each array point. Two density models are used for the gas flow out of a single capillary tube.

(i) Cone model. The gas flow is assumed to have a cone shape of which the top angle can be specified. Array points within the cone are assigned a weight proportional to the inverse square of the distance from the array point to the exit of the capillary tube. Array points outside the cone are rejected.

(ii) Model of Olander and Kruger (1970). For gas flow through a single capillary tube Olander and Kruger (1970) showed that the gas density outside the tube is given by

$$n(x, y, z) = \frac{N}{V} \frac{j(\alpha)}{\pi r^2} \quad (4.4.16)$$

where $N$ is the total flow rate (number of particles per second) through the tube, $V = \sqrt{3kT/\mu}$ is the average thermal velocity of particles in the jet, $j(\alpha)$ is a normalised distribution function that depends on flow conditions in the tube and is typically taken as $\cos \alpha$ or $1$, $\alpha$ is the angle between the direction to the array point and the
centerline of the tube, \( r \) is the distance from the tube exit to the array point, and \( K \) is the transmission probability (Clausing factor) of the tube. Array points are assigned a weight proportional to the density given by this equation.

A second weight factor is introduced to account for the variation of the differential cross section. For the noble gases the differential cross section often decreases over several orders of magnitude when the scattering angle increases from 0 to 30 degrees. Within the small range of scattering angles accepted by the electron analyser the smaller scattering angles will be slightly favoured. This effect was found to be significant in the forward scattering studies of Martus et al (1988). For the heavier rare gases the program uses the theoretical values of the differential cross sections calculated by Bartschat and Madison (1987) and for helium the calculations of Madison and Winters (1983) are used.

The finite acceptance angles of the photon channeltron and the electron analyser are implemented in the program as options. Effects due to these are calculated by choosing a few grid points within the radius of the channeltron cone or within the radius of the entrance aperture of the electron analyser. For each array point the signals are obtained by adding the intensities calculated individually for each grid point.

Variation in the transmission function of the electron analyser was taken account of by assuming that this function was triangular in shape. This amounted to weighting grid points overlaid on the entrance aperture of the electron analyser according to their distances from the center of the aperture.

In summary the numerical model program simulated the following effects:
(i) the finite radius, displacement, and convergence/divergence of the electron beam,
(ii) the finite dimension, displacement, and density profile of the gas jet,
(iii) the variation of the Stokes parameters and the differential cross section as a function of the scattering angle within the interaction region,

(iv) the optical properties and finite acceptance angle of the $P$, photon detector, and

(v) the finite acceptance angle of the electron analyser.

A detailed discussion of the results obtained using the different models is presented in Chapter VI. A paper based on this analysis (van der Burgt et al 1990) has been submitted to J. Phys. B.
Chapter V

APPARATUS AND EXPERIMENTAL CONSIDERATIONS

5.1 An Overview of the Experiment

The basic experimental arrangement is shown schematically in Figure 8. Electron impact excitation of the atomic species being studied was accomplished with the intersection of an approximately monoenergetic electron beam with an atomic gas jet, thereby defining the interaction region. The exciting electrons were scattered into the electron detection system, which, in part, consisted of an entrance aperture followed by a hemispherical electron energy analyser, which could be positioned to accept electrons scattered through a chosen angle, \( \theta \), and a channel electron multiplier (or "CEM", or "channeltron") upon which electrons impinged and were counted. The hemispherical analyser was tuned to accept only electrons which had lost an amount of energy corresponding to the excitation energy of the atomic state of interest. After traversing the analyser the electrons were accelerated such that their energy was sufficient for their detection upon striking the CEM.

VUV photons emitted from the interaction region, due to the decay of the excited atomic state, were detected in one of two directions, both perpendicular to the incident electron beam axis (the x-axis). Photon detection included polarisation analysis by means of single or double reflection polarisers followed by channeltrons. The out-of-plane double reflection polarisation analyser was used to measure the linear polarisations \( P_1 \) and \( P_2 \) and the circular polarisation \( P_3 \), while the in-plane single reflection polarisation analyser was employed only for \( P_4 \) measurements.

Signals from the electron and photon channeltrons were routed through standard coincidence circuitry to be accumulated by a multichannel analyser (MCA) working in pulse height analysis (PHA) mode. As discussed in Section 3.3, Stokes parameter
Figure 8. Schematic of the apparatus.

(or polarisation) measurements require intensity measurements for two orientations of the polarisation analysers. To accomplish this the MCA/P HA was programmed to trigger stepper motors attached to the mirror mounts so that the polarisers were rotated by the proper amount after specific data taking periods. The signals corresponding to different polariser orientations were stored in separate memory groups of the MCA/P HA and upon completion of a data run the data was transferred to a computer for analysis.
Total data accumulation times depended upon the desired statistical accuracy of the results, which in turn was greatly limited by the electron scattering angle, with larger scattering angles requiring significantly longer accumulation times. Linear polarisation measurements could be accumulated much faster than circular polarisation measurements at the same scattering angle. Typically linear polarisation data required one day at smaller scattering angles and about one week for \( \theta_s = 50^\circ \) (the largest scattering angle used). In contrast, circular polarisation measurements ranged from three days at smaller angles to three weeks at \( \theta_s = 25^\circ \), which was the largest scattering angle for \( P_3 \) measurements. Although large angle results for all parameters would be useful it was very difficult to maintain experimental stability for one week, let alone longer time periods.

The remainder of this chapter is a detailed discussion of the various apparatus and experimental components. The first topic is a brief discussion of the vacuum system, following which, for the purpose of elucidation, the experiment is decomposed into four sections:

(1) the source region,
(2) the electron detection system,
(3) the photon detectors, and
(4) data handling and management.

5.2 The Vacuum System

The experimental chamber was maintained under high vacuum by a turbomolecular pump. Pressures in the 20" x 20" x 20" chamber were measured 10" above the bottom of the chamber, at the level of the interaction region, with an ionisation gauge. Background pressures in the chamber, usually attainable overnight, were \( \sim 3 \times 10^{-7} \) torr.
Measured pressures and any pressures quoted in this work have not been corrected for the particular gas. In an attempt to attain the lowest possible background pressures, Viton O-rings were used and the system was gently baked regularly.

A belt driven oil-lubricated pump was used to rough out the system and to exhaust the turbomolecular pump for all of the Ne, Ar, and Kr work, and for about one-third of the Xe work. It was found that oil-based pumps caused undue contamination of the gold-coated mirrors, and hence experimental instability. This problem was dealt with by a "mirror cleanliness" normalisation (which is explained in detail in Section 5.5.1), but the best solution to the problem was realised with the use of an oil-free backing pump. This pump led to extremely stable mirror conditions, after the system was relatively cleansed of oil, and improved the operation of the entire apparatus in general.

For proper operation of the experimental components within the experimental chamber it was essential that stray magnetic fields be excluded from the interior of the chamber. This was accomplished by the presence of a cube-shaped μ - metal shield inside the vacuum chamber and enclosing all experimental components. The bottom of the shield was drilled with an array of half inch diameter holes to allow proper vacuum pumping. The residual magnetic field within the shield was found to uniformly be ~ 10 milligauss, which was completely acceptable.

Additionally, a set of Helmholtz coils were mounted exterior to the vacuum chamber to help cancel residual fields. However these were found to be unnecessary and had no effect on polarisation measurements.

5.3 The Source Region

The source region consisting of the electron gun, the gas jet, and the interaction region, is displayed in Figure 9.
5.3.1 The Electron Gun

The electron gun was a five element, aperture lens design which produced approximately monoenergetic electrons by thermionic emission from a commercially available tungsten filament. The non-magnetic molybdenum lens elements were used as electrostatic focussing electrodes to focus the electron beam as desired and were held in place by ceramic rods and Macor spacers. The five lens elements permitted the gun to be used in accelerating, Einzel, or decelerating mode by electrically tying certain lens elements together, though in this work the gun was only used in the accelerating mode. Additionally (see Figure 9), there were two sets of deflector plates to help steer the electron beam. The electron gun was enclosed in an aluminum and copper housing to prevent stray electrons from the filament from reaching the interaction region.

Figure 9. Schematic of the source region. The intersection of the electron beam and the gas jet determines the interaction region.
Electrons were accelerated from the cathode, which was maintained at a negative potential, to the grounded skimming aperture and then entered the grounded interaction region. Thus, the negative of the cathode potential gave the electron impact energy, $E_e$. When the electron energy analyser was not in the way, unscattered electrons were collected in the Faraday cup, which is discussed further in Section 5.4.4.

The filament heating current was typically in the 1.9 - 2.5 Amp range, which produced electron currents in the range 0.2 - 2.0 μA as measured on the outer hemisphere (this is discussed in more detail in Section 5.4.2). As standard procedure the electron gun was focussed for maximum current on the outer hemisphere when the electron analyser was positioned at $\theta_\alpha = 0$. The lens element potentials required to achieve this agreed very well with the values given by an in-lab electron optics program based on the data of Harting and Read (1976).

Since the energy spread of the beam was a few hundred meV, complete separation of the $s\left[ 1/2 \right]_0$ and $s\left[ 3/2 \right]_0$ peaks in Ne and Ar was not attainable. The electron energy scale was calibrated by measuring the threshold excitation of the 58.4 nm ($2^1P$) line of helium with its onset at 21.22 eV.

5.3.2 The Gas Jet

The gas jet emanated from a single capillary non-magnetic molybdenum tube of inner diameter 0.5 mm, the end of which was located about 2 mm from the center axis of the electron beam. There were two gas needles, one oriented at 45° above the scattering plane and at 45° from the $x - z$ plane (as shown in Figure 9), and another situated perpendicular to the electron beam and pointing vertically upward. For the vast majority of this work the 45° needle was employed. The 90° needle was installed during investigations of the effect of the finite volume of the interaction region since
it provided a much more symmetrical arrangement. Careful and extensive testing showed that the measured Stokes parameters were independent of which gas jet orientation was used.

Experiments were performed using research grade gases with stated purities of at least 99.995%. Gases were leaked into the interaction region such that the background pressure increased less than $2 \times 10^7$ torr in order to render negligible any radiation trapping effects. For similar background pressure increases, different head pressures, measured in the gas lines, were required for each gas. For a typical background pressure increase of $1.5 \times 10^7$ torr the head pressures ranged from $\sim 28$ mtorr for Xe to $\sim 250$ mtorr for Ne.

For each gas used, and for He, non-coincident polarisation measurements were taken as a function of background pressure increase and absolutely no pressure dependence was found until background pressures increased by two orders of magnitude beyond the typical increases used during the normal Stokes parameter measurements. Similarly increasing these pressures by an order of magnitude had no effect on Stokes parameter measurements.

Each gas needle was electrically isolated from all other parts of the interaction region and was held at target potential, allowing currents to be measured on them. This gave a good indication of the quality of the electron beam, and it was always found that essentially no current was picked up on the gas needles.

5.3.3 The Interaction Region

As indicated in Figure 9, the interaction region was surrounded by a grounded rigid wire cage to prevent any electrical field penetration into the interaction region. The skimming aperture of the electron gun and the gas needles protruded into the cage.
The entire "front" of the electron gun facing the interaction region, the gas needles, the grounded cage, the analyser nose cone, and the Faraday cup were all sooted to prevent reflections and secondary electron emission.

Every time any maintenance was performed on the electron gun or the gas needles, an alignment pin was inserted in the front of the electron gun and the alignment of the electron beam axis and the electron analyser position \( 0_0 = 0 \) was verified.

### 5.4 The Electron Detection System

The electron detection system comprised the electron energy analyser entrance aperture, the hemispherical electron energy analyser, the electron channeltron, and the Faraday cup. A schematic of this system is shown in Figure 10.

#### 5.4.1 The Entrance Aperture

The analyser entrance aperture and nose cone are the components of the electron detection system which protruded furthest toward the interaction region and hence, when \( 0_0 = 0 \), gave details concerning the performance of the electron beam. The stainless steel nose cone, with its front aperture inner diameter of 2 mm, was held at target potential and was electrically isolated from the molybdenum 1 mm diameter entrance aperture (see Figure 10), which was also at target potential and was the first lens element of a three element lens stack whose purpose was to focus the scattered electron beam into the electron energy analyser. Currents could be measured on both the nose cone and the entrance aperture, and currents that were essentially zero, for \( 0_0 = 0 \), indicated good alignment and small spreading of the electron beam.

The lens stack only possessed one variable potential lens element since the analyser entrance aperture was maintained at target potential and the final lens element, the hemisphere aperture, was matched to the electron analysing energy (ex-
Figure 10. The electron detection system. The system components are the entrance aperture, the hemispherical electron energy analyser, the electron channeltron, and the Faraday cup.

explained in the next section). Even with only one adjustable lens element and a set of defectors the tuning of the lens stack had very substantial effect on the detected electron count rate. At smaller scattering angles the nose cone and/or entrance aperture often eventually charged up and prevented some of the scattered electrons from entering the electron detection system. This is problem enough, but if it is not known that charging has occurred the lens stack might be tuned such that scattered electrons from away from the nominal scattering center are preferentially accepted into the detector. Then the problem is not just that of a reduced coincidence rate, but
depolarisation as a result of a possibly unnoticed experimental effect as well. Once charging was detected the system was opened and the nose cone and analyser were abrasively cleaned and resooted.

A number of experiments with controlled defocussing of the lens stack yielded dramatic depolarisations, indicating that it is essential to detect only electrons from the nominal scattering center.

### 5.4.2 The Hemispherical Electron Energy Analyser

The 180° hemispherical energy analyser employs an inverse-square-law field created by placing a potential across a pair of concentric hemispherical electrodes. The field between the hemispheres is given by

\[
\overline{E}(r) = \frac{(V_1 - V_2)}{r^2} \frac{R_1 R_2}{(R_2 - R_1)} r
\]  

where \( V_1 \) and \( V_2 \) are the potentials on hemispheres \( R_1 \) and \( R_2 \), respectively. If the incoming electron beam has energy \( E \) and enters the hemispheres at a radius \( R_o \) given by

\[
R_o = \frac{R_1 + R_2}{2}.
\]

then the potentials required for transmission of the electrons, on a circular orbit, are

\[
V_1 = E \left[ 2 \frac{R_o}{R_1} - 1 \right]
\]

and

\[
V_2 = E \left[ 2 \frac{R_o}{R_2} - 1 \right].
\]

From the preceding two equations the transmitted energy, called the analysing energy, can be found:
\[ E = \frac{1}{2R_0} \left( \frac{R_1 R_2}{R_2 - R_1} \right)^{\frac{1}{2}}. \]  

(5.4.2.5)

In this work the hemisphere dimensions were such that \( R_0 = 2R_1 \) and therefore \( R_0 = 3R_1/2 \). Equation (5.4.2.5) then has the simplified form:

\[ E = \frac{2}{3} \left( 1 - \frac{1}{3} \right)^{\frac{1}{2}}. \]  

(5.4.2.6)

It is desirable to choose \( l_1 \) and \( l_2 \) close to the analysing energy to minimise distortion of the field in the gaps between the hemispheres.

The resolution of the 180° analyser is given by

\[ \frac{\Delta E}{E} = \frac{d}{2R_0} + \frac{1}{2} (\Delta \alpha)^2 \]  

(5.4.2.7)

where \( d \) is the hemisphere aperture diameter and \( \Delta \alpha \) is the maximum angular deviation of an incident trajectory with respect to the central path. In this work the hemisphere aperture diameter was 0.5 mm and the hemispheres were 2" and 4" in radius, leading to a resolution of less than 100 meV with the usual analysing energy of 15 eV. From Eq. (5.4.2.7) it is clear that resolution improves with decreasing analysing energy, but occasionally resolution was forsaken for the sake of larger count rates in the Kr and Xe cases where even poor resolution permitted separation of adjacent energy levels. Conversely, in an attempt to better separate the \( s^+[1/2]^0 \) and \( s^-[3/2]^0 \) states in Ne and Ar, the resolution was often improved by using an analysing energy of 2 eV. It should be pointed out that the overall resolution of the experiment was still determined by the energy spread of the electron gun. Also, it is remarked that the energy \( E \) is with reference to the electron detection system, as are \( l_1 \) and \( l_2 \).

Hemispherical energy selectors suffer from aberrations due to fringe fields in the gap between the hemispheres. The fringe fields are due to the fact that half of each sphere is missing. In order to achieve the proper inverse-square field dependence
around the mean orbital radius of the electron beam, the potentials of the apertures at the entrance and exit of the hemispheres were set to match the analysing energy and variable potential correcting hoops were employed (see Figure 10). Hoop potentials were applied to minimise distortion of the field at the entrance and exit of hemispheres (see Brunt et al 1977).

A deflector in the horizontal plane through the hemispheres (the $x - y$ plane of the scattering symmetry) allowed trajectory adjustment and had an enormous effect on electron detection rates.

The purpose of the energy analyser was to transmit only electrons which had lost an amount of energy corresponding to the excitation energy of the state of interest. This was realised by setting the analyser ground to be equal to the impact energy minus the energy loss. Then only electrons which had lost energy corresponding to the difference between the electron gun ground and the electron detector ground would reach the electron channeltron.

Energy loss could be varied by adding an increasing ramp voltage from the MCA/PHA to the detector ground resulting in an energy loss that decreased as the ramp voltage increased. This gave an energy loss spectrum for a given impact energy. Energy loss spectra were, in general, simple measurements and hence were taken quite frequently to allow continual monitoring of the experiment. Energy loss spectra were particularly useful for revealing any resolution problems or other problems which might have caused the experiment to drift off of the desired energy loss peak for the state of interest.

As mentioned in Section 5.3.1, the electron beam was focussed for maximum current on the outer hemisphere, $S_2$. This was performed after positioning the analyser at $\theta_v = 0^\circ$ and then setting the inner hemisphere potential to zero so that electrons would be attracted to the outer hemisphere only. It was found that this method was
independent of whether or not the energy loss was set to zero. Current could be measured on the hemisphere aperture, allowing monitoring of the scattered electron beam. Measurements of current on S2 as a function of analyser angle $\theta$, yielded a very symmetric electron beam profile centered at $\theta = 0^\circ$ with a full-width at half-maximum of 2.4$^\circ$.

5.4.3 The Electron Channeltron

After energy selected scattered electrons exited the hemispheres they passed through a three element lens stack which focussed them into the electron channeltron where they were counted. The first lens element of the stack was the hemisphere exit aperture, which was matched to the electron analysing energy, and hence only the latter two elements and a set of deflectors were available for focussing the electrons for maximum count rate. Current could be measured on the hemisphere exit aperture allowing alignment of the energy analysed scattered electron beam.

The Galileo Electro-Optics model 4039C channeltron was mounted in a two-piece housing with the outer housing maintained at target potential and the inner housing delivering the CEM cone bias. The cone bias was typically kept at about +100 V with respect to the last lens element of the lens stack. This served two purposes: (1) electrons were attracted toward the channeltron cone, and (2) the electrons were accelerated such that their energy easily exceeded the ≈8 eV work function of the channeltron cone surface.

A very stable Tennelec high voltage supply delivered 3 - 5 kV to the spiral tail of the channeltron to facilitate the necessary electron avalanche effect, and signal pulses were routed to the counting electronics. All channeltrons in the experiment were run in a saturated condition, meaning that increasing the tail high voltage did not increase the count rate. Since channeltron lifetime depends on total signal detected over the
period the CEM is in operation, an upper limit to the electron count rate was established. In order to extend the life of the CEM but still achieve a reasonable count rate, the electron beam current was adjusted such that the electron CEM was usually operating at a count rate of less than 10 kHz.

5.4.4 The Faraday Cup

The most serious persistent problem with the apparatus was associated with stray electrons leading to spurious counts in both the electron and photon channels. For this reason, as in most experiments, a Faraday cup was used to collect the electron beam. The Faraday cup consisted of an outer housing at target potential and an inner collecting electrode at about +200 V with respect to target. Currents could be measured on both elements and it was found that beyond a collecting potential of $\sim 50$ V, collected current on the inner Faraday cup did not depend on its potential. The collecting electrode possessed a cone shaped base so that electrons scattered from it would be deflected toward the housing and therefore would be less likely to escape back toward the interaction region.

It was required that the Faraday cup could be moved out of the way of the electron detector entrance aperture when it was positioned at small scattering angles. For this purpose an adjustable teflon spacing screw and a spring system were utilised to keep the analyser nose cone and the outer Faraday cup very close to each other, but not in contact, while allowing the Faraday cup to remain in line with the electron beam once the analyser went beyond $\theta_s \sim 28^\circ$. The outer Faraday cup was manufactured such that it did not extend as far toward the interaction region as the analyser nose cone did. This was to prevent electrons from scattering off the Faraday cup and into the electron analyser. Although the Faraday cup was not completely in position until $\sim 28^\circ$, most of the electron beam was collected in the Faraday cup when the analyser was at $\theta_s \sim 12^\circ$. 

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5.5 The Photon Detectors

The photon detectors consisted of one or two polarisation analysers (or polarisers), which were gold-coated mirrors positioned for a particular angle of incidence, and a channeltron. Figure 11 shows a schematic diagram of the photon detectors. For P\(_1\) measurements only one reflection was employed and the situation may be visualised by ignoring the reflector labelled R\(_1\).

5.5.1 The Polarisation Analysers

![Diagram of photon detectors](image)

**Figure 11. The photon detectors.** For P\(_1\) measurements only one reflection is employed and hence reflector R\(_1\) may be ignored.

The theory of the VUV reflection polarisation analyser has been detailed by Westerveld *et al* (1985) and the reader is referred there for a full description. The present analysers employed gold-coated optics with an angle of incidence of \(\theta_1 = 57.5^\circ\).
For the double reflection polariser this meant that the second reflector was positioned such that its angle of incidence was also $\theta_r = 57.5^\circ$. For 58.4 nm radiation this angle of incidence produces a phase shift, between the two orthogonal components of the electric field, of $\pi/2$ in the reflected beam. At the wavelengths under consideration in this work (70 - 150 nm), the phase shift is close to, but not equal to, $\pi/2$. As a result, after one reflection any originally circularly polarised radiation becomes nearly completely linearly polarised. In this sense its performance is analogous to that of a 1/4-wave plate in transmission polarisation measurements.

For in-plane measurements the single reflection polariser was used and was oriented such that it detected radiation polarised parallel to or perpendicular to the scattering plane and, as discussed in Section 2.2, the $P_1$ linear polarisation parameter determined. For $P_1$ and $P_2$ linear polarisation measurements the out-of-plane double reflection polariser was employed. With the mirror normal vectors lying in the same plane the double reflection polariser acted as a double linear polariser with a higher degree of final linear polarisation. Orienting this plane to be parallel to or perpendicular to the incident electron beam (the x-axis) allowed the $P_1$ linear Stokes parameter to be measured. With an orientation different by 45° the $P_2$ linear Stokes parameter could be measured. An in-vacuum stepper motor permitted the second mirror to be rotated around the direction defined by the reflected beam from the first mirror.

The situation was somewhat different for $P_3$ circular polarisation measurements. Once $P_1$ and $P_2$ were known for a particular scattering angle and impact energy for the state of interest, the charge cloud alignment angle, $\gamma$, was determined via Eq. (3.7.31). The alignment angle, $\phi^*$, of the polarisation ellipse was shown by Westerveld et al. (1985) to be

$$\phi^* = \frac{1}{2} \tan^{-1} \left( \frac{P_1}{P_2} \right) .$$  \hspace{1cm} (5.5.1.1)

Comparing $\phi^*$ to $\gamma$, the orientation of the polarisation ellipse is given by
\[ \phi' = \gamma - \frac{\pi}{4} \quad (5.3.1') \]

Setting the plane of incidence of the first mirror to \( \phi' \) with respect to the \( x \)-axis then allowed circular polarisation measurements to be obtained by measuring intensities with second mirror plane of incidence settings of \( \pm \pi / 4 \) with respect to the polarisation ellipse orientation \( \phi' \).

For a detector assumed to be polarisation insensitive and for a reflection phase difference assumed to be \( \pi / 2 \), Westerveld et al (1985) showed that the circular polarisation is given by

\[ P_3 = \frac{(R_z + R_p)^2}{2(R_z - R_p)(R_z R_p)^{1/2}} \left( \frac{l\left( \frac{n}{4} \right) - l\left(- \frac{n}{4} \right)}{l\left( \frac{n}{4} \right) + l\left(- \frac{n}{4} \right)} \right) \quad (5.3.1.3) \]

where \( l(\pm \pi / 4) \) are intensities measured at \( \pm \pi / 4 \) with respect to polarisation ellipse orientation \( \phi' \). The reflection coefficient term gives the polarisation sensitivity of the device. In this work the phase shift between the orthogonal components of the photon electric vector was never quite \( \pi / 2 \). Khakoo and McConkey (1987) have shown that taking non-\( \pi / 2 \) phase shifts into account gives

\[ P_3 = \frac{(R_z + R_p)^2}{2(R_z - R_p)(R_z R_p)^{1/2} \sin \Delta_r} \left( \frac{l\left( \frac{n}{4} \right) - l\left(- \frac{n}{4} \right)}{l\left( \frac{n}{4} \right) + l\left(- \frac{n}{4} \right)} \right) = P_{\text{rot}} \quad (5.3.1.4) \]

where \( \Delta_r \) is the phase shift. Here the + sign is taken if \( \phi' = \gamma - \frac{\pi}{4} \) is used and the - sign is taken if \( \phi' = \gamma + \frac{\pi}{4} \) is used for experimental convenience.

As well, as discussed in section 4.4, the polarisation efficiency of the optical components (i.e. the mirrors) must be taken into account for linear polarisation measurements. All of the equations in the theoretical treatment in Chapter III, and in most of Chapter IV, related the Stokes parameters to intensity measurements assuming a polarisation efficiency of unity for the devices, but in practice the efficiencies are
less than unity. Analogous to Eq. (4.4.13), the polarisation efficiency for a device employing \( n \) reflections with the same angle of incidence is given by

\[
\eta = \frac{R_n^p - R_n^s}{R_n^p + R_n^s}, \quad (5.5.1.5)
\]

and the Stokes parameters are related to the measured intensities by

\[
P_i = \frac{1}{\eta} \frac{l(0^\circ) - l(90^\circ)}{l(0^\circ) + l(90^\circ)}, \quad i = 1, 2, 3, \quad (5.5.1.6)
\]

Lynch and Hunter (1985) reported extensive experimental \( n \) and \( k \) values, for gold surfaces, as functions of wavelength. Their data was used in the formulation of Samson (1967) to calculate \( R_s \) and \( R_p \) values, and hence polarisation efficiency values, \( \eta \). Inverse polarisation efficiencies, \((1/\eta)\), for single gold mirrors are plotted as a function of wavelength in Figure 12. Inverse polarisation efficiencies at wavelengths of interest to this work are shown for both single and double reflection polarisers in Table 2.

As a continual check of the experiment, polarisation ellipses were measured by accumulating non-coincident measurements of \( l(0^\circ) \), \( l(90^\circ) \), \( l(180^\circ) \), and \( l(270^\circ) \), as they related to \( P_1 \) and \( P_4 \) measurements, before and after every Stokes parameter measurement. The experiment was operating properly if intensities 180° apart were similar. These same measured quantities were also used for the previously mentioned "mirror cleanliness" normalisation. The normalisation involved first calculating the non-coincident polarisations

\[
P_{nc} = \frac{1}{\eta} \frac{[l(0^\circ)+l(180^\circ)] - [l(90^\circ)+l(270^\circ)]}{[l(0^\circ)+l(180^\circ)] + [l(90^\circ)+l(270^\circ)]}, \quad (5.5.1.7)
\]

both before and after the Stokes parameter measurement, and then taking a weighted mean of the two to give the non-coincident polarisation for the data set. Since pola-
risation efficiency decreases as mirror contamination increases, the measured Stokes parameters were scaled up by the normalisation factor

\[ \kappa = \frac{P_h}{P_{ac}} \]  

(5.5.1.8)

Figure 12. Single mirror inverse polarisation efficiencies. Inverse polarisation efficiencies for single gold mirrors are plotted as a function of wavelength. Based on the data of Lynch and Hunter (1985), which did not include error bars.

where \( P_h \) was the highest measured non-coincident polarisation for a particular species at a particular impact energy. In practice the normalisation factors for \( P_1 \) (which were also valid for \( P_2 \) and \( P_3 \) since they used the same double polariser) were always very close to unity, while the correction factors for \( P_4 \) were more significant. This was to be expected since Eq. (5.5.1.5) shows that double reflection polarisers should be consid-
Table 2. Inverse polarisation efficiencies of the gold mirrors. Inverse efficiencies have been calculated from the data of Lynch and Hunter (1985).

<table>
<thead>
<tr>
<th>Species</th>
<th>State</th>
<th>λ (nm)</th>
<th>Energy (eV)</th>
<th>$\frac{1}{\eta}$ (1 refl.)</th>
<th>$\frac{1}{\eta}$ (2 refl.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>$2^1P$</td>
<td>58.4</td>
<td>21.22</td>
<td>1.536</td>
<td>1.094</td>
</tr>
<tr>
<td>Ne</td>
<td>$3^1P$</td>
<td>73.6</td>
<td>16.85</td>
<td>1.479</td>
<td>1.077</td>
</tr>
<tr>
<td></td>
<td>$3^3P$</td>
<td>74.4</td>
<td>16.67</td>
<td>1.487</td>
<td>1.080</td>
</tr>
<tr>
<td>Ar</td>
<td>$4^1P$</td>
<td>104.8</td>
<td>11.83</td>
<td>1.412</td>
<td>1.060</td>
</tr>
<tr>
<td></td>
<td>$4^3P$</td>
<td>106.7</td>
<td>11.62</td>
<td>1.419</td>
<td>1.062</td>
</tr>
<tr>
<td>Kr</td>
<td>$5^1P$</td>
<td>116.5</td>
<td>10.64</td>
<td>1.442</td>
<td>1.068</td>
</tr>
<tr>
<td></td>
<td>$5^3P$</td>
<td>123.6</td>
<td>10.03</td>
<td>1.451</td>
<td>1.070</td>
</tr>
<tr>
<td>Xe</td>
<td>$6^1P$</td>
<td>129.6</td>
<td>9.57</td>
<td>1.457</td>
<td>1.072</td>
</tr>
<tr>
<td></td>
<td>$6^3P$</td>
<td>147.0</td>
<td>8.44</td>
<td>1.458</td>
<td>1.072</td>
</tr>
</tbody>
</table>

erably less affected by small variations in mirror surface quality. Consequently, this correction factor was only used for $P_\star$ measurements in the final analysis. Over the entire course of this work $P_\star$ normalisation factors varied between 1.004 and 1.226. Once the oil-free backing pump was installed the deviations of $P_\star$ non-coincident measurements from the high value were typically about 2.5%. The $P_\star$ mirror was continually heated to about 70°C to reduce vapour condensation on its surface and improve cleanliness. Maximum measured $P_\star$ non-coincident polarisation values for each species are shown in Table 3. Note that these measurements no longer relate to a particular state of the atomic species, but relate to total measured VUV radiation.
Table 3. Maximum non-coincident $P_1$ polarisations.

<table>
<thead>
<tr>
<th>Species</th>
<th>Impact Energy (eV)</th>
<th>Maximum non-coincident $P_1$ polarisation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ne</td>
<td>80</td>
<td>0.245 ± 0.002</td>
</tr>
<tr>
<td>Ar</td>
<td>80</td>
<td>0.128 ± 0.005</td>
</tr>
<tr>
<td>Kr</td>
<td>60</td>
<td>0.192 ± 0.002</td>
</tr>
<tr>
<td>Xe</td>
<td>30</td>
<td>0.148 ± 0.002</td>
</tr>
<tr>
<td></td>
<td>50</td>
<td>0.160 ± 0.002</td>
</tr>
<tr>
<td></td>
<td>80</td>
<td>0.115 ± 0.003</td>
</tr>
</tbody>
</table>

Other periodic tests of the experiment were $P_1$ Stokes parameter measurements for He ($2^3P_1$). If measured $P_1$ values were not unity then a problem existed in the experiment.

5.5.2 The Photon Channeltrons

The channeltrons used for photon detection were identical to the electron channeltron. Unfortunately, the quantum efficiency for photon detection with a CEM is only ~10% up to 100 nm and then decreases rapidly with increasing photon wavelength (see, for example, Weller and Young 1970). Johnson (1969) demonstrated that coating the entrance cone of channeltrons with cesium iodide enhanced their detection efficiency by about two orders of magnitude at wavelengths above 100 nm in the VUV. Since the Ar, Kr, and Xe transitions of interest in this work lie above 100 nm, this technique was employed, resulting (according to Johnson 1969) in detection efficiencies of ~10% even at the longest wavelength considered in this work.
Physically, the photon channeltrons were mounted in grounded housings attached rigidly to the mirror mounts meaning that rotation of a polariser was actually a mirror and a CEM moving as a unit. For the double reflection polariser the CEM and the second mirror were a unit which moved independently of the first mirror.

High voltages applied to the photon channeltrons were of similar size to those applied to the electron CEM, but the count rate rarely reached 4 kHz and hence the lifetimes of these channeltrons were substantially longer than that of the electron CEM. The nose cones of the photon channeltrons were biased about -200 V with respect to target to prevent stray electrons from entering the photon detection system. At biases near this level the photon signal was independent of bias potential.

5.6 Data Accumulation and Management

Polarisation measurements required intensity measurements for two orientations of the polariser in use (only one polariser could be used at a time). Polariser orientation was changed by programming the Tracor Northern TN-1710 MCA/PHA to trigger the stepper motor attached to the appropriate mirror mount. Data for each orientation was stored in a separate memory group of the Tracor Northern, along with its total accumulation time. Accumulation time at each orientation was preset according to the expected polarisation since this quantity indicated which intensity should be weaker and, in the quest for better statistics, programming was such that data was accumulated proportionally longer for the weaker intensity. A complete cycle of polariser orientations took 10 minutes, and several hundred such cycles were performed during a data run to average out the effects of any minor experimental instabilities.
Signals from the electron and photon channeltrons were routed into the fast coincidence electronics to be processed, so that a coincidence spectrum for each polariser orientation could be accumulated. The next section details the operation of the coincidence electronics.

5.6.1 The Coincidence Electronics

Figure 13 shows a schematic diagram of the counting electronics. Signal pulses from the electron and photon channeltrons (typically -20 mV, 20 nsec) were sent through $\times 10$ preamplifiers enroute to timing filter amplifiers (TFAs), which were used to amplify the pulses to -1 V. The TFAs were also used for pulse shaping, which allowed experimental artifact or noise features to be somewhat controlled. The TFA outputs were fed into constant fraction discriminators (CFDs) which were set to discriminate against pulses of magnitude less than 0.5 V. The positive outputs of the CFDs allowed rate meters to give an analogue indication of the count rates.

The negative -0.9 V CFD outputs went on different routes for the electron and photon pulses. The photon CFD negative output simply provided a start pulse for a time-to-amplitude converter (TAC). The electron CFD negative output was routed through another TFA and then delayed, relative to the photon pulse, before providing a stop pulse for the TAC. This second TFA was required for amplification purposes since the signal coming out of the delay generator was usually greatly reduced in size. The adjustable delay allowed the position of the coincidence peak to be varied in the final spectrum.

The TAC converted the time between start and stop pulses into analogue pulse heights which were then converted into digital signals by the analogue-to-digital converter (ADC) of the pulse height analyser (PHA) of the Tracor Northern. The digital signals were then analysed by the multi-channel analyser (MCA) of the Tracor
Figure 13. The coincidence electronics.

Northern to generate a time spectrum. Electron-photon pairs originating from the same collision event appeared as a coincidence peak superimposed on a flat background in the time spectrum. A sample coincidence spectrum is shown in Figure 14.

To measure non-coincident polarisations start pulses for the TAC were still obtained from the photon channel, but true starts from the TAC, rather than the converted signal, were sent to the MCA where they were simply accumulated. Electron pulses were completely disregarded for these measurements.

To measure energy loss spectra the PHA module of the Tracor Northern was replaced with a single channel analyser (SCA) module. In this mode a positive 0 - 5 V ramp output from the Tracor Northern was applied against the electron detector potential resulting in a varying energy loss. The energy loss spectrum resolution was
Figure 14. A sample coincidence spectrum. The coincidence peak occurred at about 500 nsec for the usual delays applied in the electron channel.

given, roughly, by the 5 V ramp divided by the number of storage channels in the memory group. The positive output pulses of the electron CFD allowed a time spectrum to be measured for each channel in the memory group. Each time spectrum was measured during a preset dwell time and the total count sent to be stored in the appropriate memory address of the MCA. The energy loss was then incremented by the proper amount and the process repeated. A sample energy loss spectrum for Xe with $\theta_e = 13^\circ$ and a constant impact energy of 50 eV is shown in Figure 15.
Figure 15. A sample energy loss spectrum. The spectrum is for Xe with \( \theta_e = 13^\circ \) and an electron impact energy of 50 eV.

5.6.2 Data Management

Data stored in the memory of the Tracor Northern MCA/PHA were transferred, via an RS232 output, to a computer where they were analysed with specially written programs.

As discussed in the previous section, a coincidence measurement consisted of a time spectrum with a coincidence peak superimposed on a flat background. A coincidence data set consisted of two time spectra: one for each orientation of the polariser. For each time spectra in the data set a *peak region* was selected which was more than sufficiently wide to include the coincidence peak. The remaining channels then constituted background and the average background per channel was calculated. This quantity was subtracted from each channel in the coincidence peak and the results
integrated to give the total intensity for that particular polariser orientation. Stokes parameters were then evaluated using Eq. (5.5.1.6) for \( P_1 \), \( P_2 \) or \( P_3 \) or using Eq. (5.5.1.4) for \( P_1 \).

For every Stokes parameter measurement the mirror cleanliness normalisation was implemented. This involved taking a set of non-coincident intensity measurements before and after the Stokes parameter data. Each set of non-coincident data included the intensities \( I(0^\circ) \), \( I(90^\circ) \), \( I(180^\circ) \), and \( I(270^\circ) \), as discussed in Section 5.5.1. These quantities were measured twice: once with normal gas flow through the gas needle, and then with the gas, instead, effusing into the chamber through a 1/4 inch orifice in the chamber wall. The first data set was considered to be signal plus background, and the second set, background. Non-coincidence polarisations were determined by subtracting appropriate backgrounds from signal plus backgrounds. The correction factor for the Stokes parameter measurement was calculated from Eq. (5.5.1.8) and was applied to obtain, finally, the measured Stokes parameter \( P_i^M \). Most of the \( P_i^M \) quoted in the results of this work are the weighted mean of many, shorter time length, individual measurements which were easier, in practice, to obtain.

The next step was to apply the analytical volume effect model to the measured \( P_i^M \) via Equations (4.3.14)-(4.3.16) to obtain the Stokes parameters \( P_i \). If no hyperfine effects were present, these \( P_i \) were the final experimental values for the Stokes parameters describing the nascent charge cloud. Otherwise, hyperfine effects were taken into account with Equations (3.7.5)-(3.7.7) and the final experimental \( P_i \) produced. To properly allow for hyperfine depolarisation effects the naturally occurring percentages of odd isotopes given by Heath (1984) were assumed. These percentages and the appropriate nuclear spins are included in Table 4. Only in Kr and Xe is any effect expected. Because the natural widths of these levels are only about 300 MHz (lifetimes of a few nsec for both Kr and Xe (Matthias et al 1977)), whereas the hyperfine
splitting is hundreds or thousands of MHz (Lederer and Shirley 1978, Husson et al 1979, Jackson 1977, Jackson and Coulombe 1972) there is more than adequate time for the hyperfine interaction to occur before emission of a photon takes place. Thus in both these targets a significant depolarisation is expected. Table 5 shows the natural widths, \( \frac{1}{\tau} \), of the Kr and Xe levels and the hyperfine splittings.

After these various corrections to the data had been performed coherence parameters such as \( \rho_{00} \) or \( \gamma \) were readily calculated.

The errors given in this work are statistical in nature and correspond to one standard deviation. The errors associated with the correction factors are small and difficult to estimate accurately and therefore have not been incorporated into the cited errors. The uncertainty in \( \theta_e \) is estimated to be \( \pm 1.5^\circ \) and the quoted energies are accurate to \( \pm 1 \text{ eV} \).
Table 4. Isotope data for the heavy rare gases.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>% Abundance</th>
<th>Nuclear Spin</th>
<th>Depolarising factor</th>
<th>Net G Factors*</th>
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<tr>
<td></td>
<td></td>
<td></td>
<td>G₁</td>
<td>G₂</td>
</tr>
<tr>
<td>Kr⁷⁷</td>
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<td>0.247</td>
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<td>Xe¹³⁶</td>
<td>8.9</td>
<td>0</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

* Ne has only a 0.257% odd isotope component. Since the effects of this will be negligible the data is not listed. Naturally occurring Ar isotopes all have zero nuclear spin.

+ These are the weighted averages of the individual G factors.
Table 5. Kr and Xe natural level widths and hyperfine splittings.

<table>
<thead>
<tr>
<th>State</th>
<th>Isotope</th>
<th>$1/\tau^a$ (MHz)</th>
<th>Hyperfine Splitting (MHz)</th>
<th>F Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kr($^1P_1$)</td>
<td>Kr$^{83}$</td>
<td>322</td>
<td>2226$^b$</td>
<td>7/2 - 9/2</td>
</tr>
<tr>
<td>Kr($^3P_1$)</td>
<td>Kr$^{83}$</td>
<td>314</td>
<td>2898$^b$</td>
<td>9/2 - 11/2</td>
</tr>
<tr>
<td>Xe($^3P_1$)</td>
<td>Xe$^{129}$</td>
<td>289</td>
<td>419$^b$</td>
<td>7/2 - 9/2</td>
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<td></td>
<td>Xe$^{131}$</td>
<td>289</td>
<td>681$^b$</td>
<td>9/2 - 11/2</td>
</tr>
</tbody>
</table>


$^b$ from Jackson (1977).

$^c$ from Jackson and Coulombe (1972).
Chapter VI
RESULTS AND DISCUSSION

6.1 Introduction

The results of this work are best presented by dividing them into three related groups: (1) results for theoretical volume effect calculations, (2) results involving measured P₁ and P₄ Stokes parameters, which will be called p₀₀ results, and (3) results involving measured P₁, P₄, and P₅ Stokes parameters, which will be called coherence results. Much of this chapter has been accepted (Corr et al 1990a, 1990b) or submitted (van der Burgt et al 1990) for publication.

Before proceeding to the results it is pointed out that the electron beam resolution of about 0.5 eV was insufficient to resolve the ns'[1/2]₀ (¹P) and ns'[3/2]₀ (³P) peaks in Ne and Ar. The Kr peaks were partially resolved. However, for the electron scattering angular range studied here the ³P peaks of Ne and Ar were approximately ten and five times the intensity of the respective ¹P peaks and so for these targets the data are predominantly due to singlet excitation even though the transitions were not resolved. In Kr the contribution from the adjacent peak was less than 10% while in Xe the two peaks were completely resolved. It is also noted that the theoretical data for the [1/2]₀ and [3/2]₀ channels are essentially identical for the scattering parameters involved here.

6.2 Theoretical Volume Effect Results

In this section data is presented to illustrate the magnitude of the volume effects discussed in Chapter IV. For the sake of clarity one data set is assumed to be accurate and it is demonstrated how this data set is modified by the experimental effects. It is assumed that the data of Bartschat and Madison (1987) are correct for the heavier rare
gases, and the data of Madison and Winters (1983) for helium. In all the data sets shown in this section this data is displayed as a solid line and the modified data are given as separate data points. The data all pertain to $I = 0$ isotopes and $Ar$ is chosen as the optimum test gas because it satisfies this restriction on nuclear spin. In the calculations the diameter of the electron detection entrance aperture was taken as 1mm and the distance from the nominal scattering center to the entrance aperture, $R_e$, was 28.5 mm.

Figure 16 shows the four Stokes parameters for $Ar^+(1/2)^0$ at 80 eV incident energy and a range of scattering angles from 0 - 30°. Here the analytical model was used for a beam radius of 1 mm and a 1 mm vertical displacement of the beam out of the nominal scattering plane. This offset required a modified version of Eq. (4.3.12). It is clear from Figure 16 that $P_{1,2,3}$ are affected in a very minor way but a serious perturbation of the $P_4$ data is evident. This is largely due to the magnifying effect of a $P_4$ which is large and negative (see Eq. (4.3.12)). Since $P_4 = 1$ in the primary data set it is clear from Eqs. (4.3.9) and (4.3.10) that modifications to $P_{1,2,3}$ will only occur at small $\theta$, where $1 - \mu = \cos^2 \beta$ diverges significantly from unity. It is noted that Simon et al (1990) also found significant interaction volume effects at small electron scattering angles, (less than 6°).

Figure 17 again presents $P_1$ and $P_4$ data for 80 eV excitation of $Ar$, but this time modified data are presented for the two models (analytical and numerical) discussed in Chapter IV. The assumed beam radius and offset are the same as in Figure 16. Very little effect is seen on the $P_1$ data; some small differences between the two models are evident in the $P_4$ data though both models show similar large deviations from the original data set at $\theta < 20^\circ$. This is an important comparison between the two models. The very similar behaviour above a scattering angle of a few degrees gives strong justification for the use of the analytical model to extract actual Stokes parameters from the measured ones.
Figure 16. Analytical model effects on Stokes parameters for Ar. Excitation is of the $^4S^-[1/2]^0_s$ state by 80 eV electrons. Solid lines indicate the DWBA data of Bartschat and Madison (1987). Note that $P_4$ coincides with unity at all angles. The squares assume an electron beam radius of 1 mm and a 1 mm vertical offset of the beam out of the scattering plane.

Figure 18 is a further presentation of the 80 eV Ar data only now the beam radius has been increased to 2 mm and the 1 mm vertical offset assumed in Figures 16 and 17 has been eliminated. A remarkable similarity between the modified data sets of Figures 17 and 18 is evident, indicating the need for careful focussing of the electron beam when coincidence measurements are attempted for small electron scattering angles.
Figure 17. Analytical - numerical model comparison. Excitation is of the $^4s\,^1[1/2]$ state by 80 eV electrons. Solid lines: Bartschat and Madison (1987), DWBA; squares: approximate analytical model; crosses: numerical model. For both models a 1 mm electron beam radius and a 1 mm vertical offset above the scattering plane have been assumed.
Figure 18. Effect of increased electron beam size and no offset. Excitation is of the $\text{Ar}^4s^2\left[\frac{1}{2}\right]^0$ state by 80 eV electrons. Solid lines: Bartschat and Madison (1987), DWBA; crosses: numerical model. A beam radius of 2 mm is assumed and no displacement of the beam from its nominal position is assumed.
Figure 19 illustrates the effect of misalignment of the electron beam in the scattering plane. The modified data shown refer to an electron beam radius of 1 mm but now offset by 1 mm from the nominal scattering center in the scattering plane. As might be expected, a more significant effect than before is observed in the $P_1$ data with a smaller but still significant effect apparent in the $P_4$ data. An effect of the same magnitude is observed in the $P_M$ data if no offset of the electron beam is assumed. Figure 19 thus shows the effect on $P_M$ of finite electron beam radius (1 mm) alone.

Figure 20 illustrates the effect of a lack of parallelism of the electron beam as it passes through the interaction region. Figure 20 shows the range of $0^\circ$, where a diverging electron beam could be significant.

Using the Olander and Kruger model (see Section 4.4) calculations were also performed in which realistic parameters were chosen for the gas jet. Within reasonable limits on these parameters very little effect on the Stokes parameters was observed indicating that the position and density profile of the gas jet are not critical. Additional calculations using the simpler cone model (Section 4.4) gave very similar results. As an experimental check of this measurements were performed using two different gas inlet systems with identical results (see Section 5.3.2).

Other factors which were considered and which showed only very minor effects were the finite acceptance angles of the photon and electron detectors and the possible variation of the optical properties of the photon detectors as they were rotated. This is not unexpected in this work because of the relatively large distances to the detectors. If however, the acceptance angles of the detectors were relatively large, proper allowance must be made for this as was done, for example, by Beijers (1987).
Figure 19. Horizontal offset of electron beam in the scattering plane. Excitation is of the $\text{Ar}^4s^+ [1/2]^0$ state by 80 eV electrons. Symbols as in Figure 18. A 1 mm radius electron beam and a horizontal offset of the electron beam of 1 mm in the scattering plane are assumed.
Figure 20. Effect of a diverging electron beam. Excitation is of the $Ar_+ s^{-3/2}$ state by 80 eV electrons. Symbols as in Figure 18. The data was modelled assuming an electron beam radius of 1 mm at the scattering center but diverging from a point 40 mm from this center. No displacement of the beam from its nominal axis was assumed.
In all calculations variation of the Stokes parameters and the differential cross section as a function of scattering angle within the interaction region were taken into account. This was only found to be significant in regions where the Stokes parameters varied rapidly with \( \theta_s \), for example close to \( \theta_s = 0 \). In this region the modified data are consistent with those of Martus and Becker (1988).

It is clear from these calculations that significant departures of \( P_4 \) from unity could be obtained if electron beam focussing and alignment were not carefully controlled. Clearly the best method to check a system for the presence of finite interaction volume effects is to use a target like Ar (or Ne) which is free from perturbing internal interactions. Any divergence of \( P_4 \) from unity could be taken as an indication of the existence of some of the effects discussed.

If it is found impossible to eliminate finite volume effects then an alternative is to include a further polarisation analyser in the scattering plane, say at 45° to the incident electron beam. This could then be used for electron scattering angles where the charge cloud has rotated to the position where it was orthogonal to the exciting electron beam and faced the original \( P_4 \) detector "end-on".

As a note of caution, however, it is pointed out that if such a detector is used then serious instrumental effects might be expected when \( P_2 \) (rather than \( P_4 \)) is large and negative. This occurs even for He under certain scattering conditions, as illustrated in Figure 21. Figure 21 shows that although these finite interaction volume effects are very small for a conventionally placed \( P_4 \) detector, they could be quite severe for one positioned at 45° to the incident beam.

The main conclusion from this study is that, provided care is taken with the focussing and alignment of the electron beam, finite volume effects are not expected to be significant except for very small angles. Measurements taken with Ne and Ar as target gases should reveal the magnitude of any effects which are occurring.
Figure 21. In-plane photon detector at 45 degrees for He. Excitation is of the $^{2}_1\!P$ state by 80 eV electrons. Solid lines: Madison and Winters (1983), DWBA; + and ×: data modelled for an in-plane photon detector at 90° and 45°, respectively, to the electron beam direction. Modelled data refer to a parallel electron beam of 1 mm radius which passes through the nominal scattering center.
6.2 $P_{m}$ Results

The data for the four target gases studied in this work are listed in Tables 7-13 and displayed in Figures 22-28. Because of the volume of this data it is presented in a page after page format starting four pages over. In all cases the displayed data are measured data. Reduced data, taking account of finite volume effects and, where applicable, hyperfine interaction effects are given in the tables. The data denoted $P_{4}^{\prime}$ are data which have been corrected, using the analytical model, for the finite volume effect via Eq. (4.3.16). The prime is simply required to indicate that these data have not undergone a correction for hyperfine depolarisation. Table 6 shows values of the parameter $\mu$ (see Section 4.3) used in this finite volume effect correction. $\mu$ was calculated from Eq. (4.3.7) with $r = 1$ mm and $R = 28.5$ mm. For comparison purposes, other available experimental and theoretical data are included in the figures. In the case of Ne, Ar, and Kr it was possible to use the $P_{1}^{\prime}$ values reported by Khakoo and McConkey (1987). A complete analysis of the $P_{1}^{\prime}$ as well as $P_{2}^{\prime}$ and $P_{3}^{\prime}$ results for these three gases was given by these authors and so the reader is referred to this paper for a full comparison of the current $P_{1}$ data with earlier work.

The $P_{1}^{M}$ values for all target gases were in good agreement with other experimental data, though very limited data was available for Ne and Ar. The best agreement with theory was with the DWBA calculations of Bartschat and Madison (1987) and so, for the sake of clarity, only these are shown on the figures. It can be seen that the agreement is quite good at the smaller scattering angles up to 20°. At larger scattering angles discrepancies are more apparent. Murray et al (1990), in their recent study of 30 eV Kr excitation, also found that the DWBA calculations were preferred.
In Kr the agreement between theory and experiment for $P^M_1$ is satisfactory for both $^5s|\frac{1}{2}\rangle^0$ and $^5s|\frac{3}{2}\rangle^0$ states though there is some indication, particularly in the latter case, that the dip in the 10° - 15° region should be somewhat deeper and broader than indicated by the DWBA or other theoretical results. Inclusion of the hyperfine depolarisation makes only a marginal improvement in the quality of the agreement.

As far as the $P^M_1$ parameter is concerned, the most significant comparisons with theory occur with Xe as the target gas. The largest angular range considered was at 30 eV impact energy, Figure 26. On Figure 26 the experimental data are compared with two theoretical curves, one excluding and one including the hyperfine depolarising effect. This figure is a clear demonstration of the significance of this effect for this target. As shown in the last section, finite volume and other effects can contribute at small scattering angles but have a negligible effect for $\theta_\circ > 20^\circ$. At the higher incident energies in Xe, Figures 27 and 28, it appears that the DWBA results again underestimate both the magnitude and width of the first dip in the $P_1$ data so that the inclusion of hyperfine depolarisation actually worsens the level of agreement between theory and experiment in this angular region. This has implications for $P_1$ data as well, as discussed shortly.

The $P^M_4$ results for Ne and Ar (Figures 22 and 23) are essentially unity over the scattering range studied here, in accord with theoretical calculations. There is the suggestion of a slight drop in the Ne $P^M_4$ values in the angular range where $P^M_1$ goes through a deep minimum, and a hint of a similar effect in the Ar data though these deviations from unity are barely statistically significant, especially if an error of two standard deviations was considered. As discussed previously, if a finite volume effect
was occurring it would be amplified for large negative values of $P_\perp$. It is therefore suggested that these data can be used to place an upper limit on the magnitude of these instrumental effects.

The Kr $P_4^H$ data are shown in the lower panels of Figures 24 and 25. Comparison is again with the theoretical data of Bartschat and Madison (1987). It is clear that the calculated curves using the theoretical data and assuming complete hyperfine depolarisation, together with the limited volume effect which arises from assuming that the electron beam has a 1 mm diameter, largely accounts for the trend in the observations. If, as suggested earlier, the theoretical data underestimate the magnitude and width of the negative excursion around $\theta_\perp = 10^\circ$, then considerably improved agreement between experiment and theory is obtained. The $5s(3/2)^1P$ data demonstrate somewhat more scatter, perhaps indicating the presence of some finite interaction volume effects in this case.

At 30 eV in Xe, Figure 26, the agreement between experiment and predictions is good over the entire angular range providing strong support for the previous conclusions that instrumental effects were small and that hyperfine depolarisation was complete, thus providing an effective "height" to the excited state charge density distribution. At 50 eV, Figure 27, the agreement is somewhat poorer but still quite satisfactory. At 80 eV the very sharp decrease in the differential cross section with $\theta_\perp$ made data taking very time consuming at the larger $\theta_\perp$ and thus data are only presented for $\theta_\perp \leq 20^\circ$. It is seen that in this case, Figure 28, very reasonable agreement is achieved between $P_4^H$ and data derived from the results of Bartschat and Madison (1987). If, as suggested earlier, the theory underestimates the negative-going excursion of $P_4$ around $\theta_\perp = 10^\circ$ then even better agreement between experimental and theoretical $P_4$ data would be obtained.
The parameters \( P_{1} \) and \( \rho_{oo} \), which are appropriate to the nascent charge cloud immediately following the original collision, have been evaluated for Ne, Ar, Kr and Xe as discussed in Section 5.6.2 and are given in the tables. Figures 29-32 show the resultant \( \rho_{oo} \) values along with theoretical predictions and the results of earlier workers. In the cases of Ne and Ar, because \( P_{1}^{M} \) was essentially unity in all of the measurements, \( \rho_{oo} \) is found to be zero within experimental error for all scattering angles, as expected.

The earlier workers performed angular correlation measurements so comparison with their results required derivations of the Stokes parameters from their \( \lambda \), \( \chi \), and \( \epsilon \) data using the transformations given in the appendices of Andersen et al (1988). Within experimental error no deviation of \( \rho_{oo} \) from zero in the angular range of \( \theta \), considered is found. This is in complete agreement with both sets of theoretical data and indicates that, in the angular range under consideration, no indication of any spin flip in the excitation process is observed. Similar conclusions were reached by Simon et al (1990) in their careful study of small angle (\(<10^\circ\)) scattering on Hg at 50 eV incident energy. They too highlighted the influence of hyperfine effects and interaction region geometry effects. The present conclusion differs from that of Plessis et al (1988) who interpreted large deviations in their \( P_{1}^{M} \) values from unity in the cases of Kr and Xe as being evidence for spin flip processes taking place. It is clear from the present work that such processes are not occurring.

It seems that the apparently non-zero \( \rho_{oo} \) values reported earlier by Danjo et al (1985) and Nishimura et al (1986) (see Figures 31 and 32) using angular correlation techniques may also be explained using similar arguments to those presented here. In Figures 31 and 32 the data from other laboratories have been corrected to take account of hyperfine interaction effects. This allows a more meaningful comparison to be made with earlier work.
Table 6. The parameter $\mu$ as a function of scattering angle. $\mu$ is calculated from Eq. (4.3.7) with $r = 1$ mm and $R_e = 28.5$ mm.

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<thead>
<tr>
<th>$\theta$ (deg)</th>
<th>$\mu$</th>
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</thead>
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<td>0.010</td>
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<td>45</td>
<td>0.001</td>
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<td>50</td>
<td>0.001</td>
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Table 7. Parameters for Ne at 80 eV incident energy.

<table>
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<th>θ,\textdegree</th>
<th>P_1^M</th>
<th>P_4^M</th>
<th>ρ_{00}</th>
<th>P_1</th>
<th>P_1^*</th>
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<td>7.5</td>
<td>-0.04±0.03*</td>
<td>0.97±0.04</td>
<td>-0.002±0.009</td>
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<td>1.01±0.04</td>
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<tr>
<td>10</td>
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<td>1.02±0.03</td>
<td>-0.010±0.006</td>
<td>-0.30±0.02</td>
<td>1.06±0.03</td>
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<tr>
<td>12.5</td>
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<td>0.98±0.06</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>-0.70±0.02</td>
<td>0.84±0.05</td>
<td>0.010±0.004</td>
<td>-0.70±0.02</td>
<td>0.88±0.05</td>
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<tr>
<td>17.5</td>
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<td>0.80±0.08</td>
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<td>-0.69±0.03</td>
<td>0.83±0.09</td>
</tr>
<tr>
<td>20</td>
<td>-0.77±0.03*</td>
<td>0.83±0.09</td>
<td>0.082±0.006</td>
<td>-0.77±0.03</td>
<td>0.87±0.09</td>
</tr>
<tr>
<td>30</td>
<td></td>
<td>0.97±0.08</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>35</td>
<td>-0.08±0.05*</td>
<td>0.99±0.06</td>
<td>0.002±0.015</td>
<td>-0.08±0.05</td>
<td>0.99±0.06</td>
</tr>
<tr>
<td>45</td>
<td>0.42±0.07*</td>
<td>1.10±0.09</td>
<td>-0.035±0.030</td>
<td>0.42±0.07</td>
<td>1.10±0.09</td>
</tr>
</tbody>
</table>

* From Khakoo and McConkey (1987).

b P_1 corrected for finite volume effect using Eq.(4.3.16).
Table 8. Parameters for Ar at 80 eV incident energy.

<table>
<thead>
<tr>
<th>$\theta_\lambda$(deg)</th>
<th>$P^M_1$</th>
<th>$P^M_4$</th>
<th>$\rho_{00}$</th>
<th>$P_1$</th>
<th>$P_{1^*}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>-0.33±0.02</td>
<td>1.00±0.02</td>
<td>-0.030±0.004</td>
<td>-0.33±0.02</td>
<td>1.19±0.03</td>
</tr>
<tr>
<td>10</td>
<td>-0.53±0.05*</td>
<td>0.91±0.07</td>
<td>0.004±0.009</td>
<td>-0.53±0.05</td>
<td>0.97±0.07</td>
</tr>
<tr>
<td>15</td>
<td>-0.76±0.04*</td>
<td>0.94±0.11</td>
<td>0.000±0.007</td>
<td>-0.76±0.04</td>
<td>1.01±0.12</td>
</tr>
<tr>
<td>20</td>
<td>-0.25±0.06</td>
<td>1.02±0.07</td>
<td>-0.005±0.013</td>
<td>-0.25±0.06</td>
<td>1.03±0.07</td>
</tr>
<tr>
<td>30</td>
<td>0.36±0.07*</td>
<td>1.04±0.08</td>
<td>-0.014±0.026</td>
<td>0.36±0.07</td>
<td>1.04±0.08</td>
</tr>
</tbody>
</table>

* From Khakoo and McConkey (1987).

Table 9. Parameters for Kr $3s^1 \; l = 1/2 \; l_1^0 \; 1P_1$ excitation at 60 eV impact.

<table>
<thead>
<tr>
<th>$\theta_\lambda$(deg)</th>
<th>$P^M_1$</th>
<th>$P^M_4$</th>
<th>$\rho_{00}$</th>
<th>$P_1$</th>
<th>$P_{1^*}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>0.81±0.05</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>-0.12±0.04</td>
<td>0.78±0.04</td>
<td>-0.001±0.012</td>
<td>-0.13±0.04</td>
<td>0.87±0.05</td>
</tr>
<tr>
<td>7.5</td>
<td></td>
<td>0.70±0.06</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>-0.77±0.06*</td>
<td>0.63±0.04</td>
<td>-0.015±0.009</td>
<td>-0.82±0.06</td>
<td>0.74±0.06</td>
</tr>
<tr>
<td>12.5</td>
<td></td>
<td>0.56±0.08</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>-0.50±0.05*</td>
<td>0.63±0.06</td>
<td>0.023±0.013</td>
<td>-0.53±0.05</td>
<td>0.65±0.06</td>
</tr>
<tr>
<td>17.5</td>
<td></td>
<td>0.78±0.07</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>-0.04±0.06*</td>
<td>0.98±0.08</td>
<td>-0.030±0.020</td>
<td>-0.04±0.06</td>
<td>0.99±0.08</td>
</tr>
<tr>
<td>30</td>
<td>0.73±0.09</td>
<td>0.96±0.05</td>
<td>-0.014±0.026</td>
<td>0.78±0.10</td>
<td>0.96±0.05</td>
</tr>
</tbody>
</table>

* From Khakoo and McConkey (1987).
Table 10. Parameters for \( \text{Kr} \uparrow s | \frac{3}{2} \uparrow | 0^+ \) \( ^3\text{P}_1 \) excitation at 60 eV impact.

<table>
<thead>
<tr>
<th>( \theta ) (deg)</th>
<th>( P_{\frac{1}{2}} )</th>
<th>( P_{\frac{3}{2}} )</th>
<th>( \rho_{00} )</th>
<th>( P_{1} )</th>
<th>( P_{1}^\ast )</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>0.04±0.04</td>
<td>0.83±0.04</td>
<td>-0.001±0.012</td>
<td>0.05±0.04</td>
<td>0.89±0.04</td>
</tr>
<tr>
<td>7.5</td>
<td></td>
<td>0.53±0.04</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>-0.71±0.03</td>
<td>0.59±0.06</td>
<td>-0.002±0.008</td>
<td>-0.75±0.03</td>
<td>0.66±0.07</td>
</tr>
<tr>
<td>12.5</td>
<td></td>
<td>0.51±0.07</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>-0.60±0.09*</td>
<td>0.66±0.06</td>
<td>0.006±0.013</td>
<td>-0.64±0.10</td>
<td>0.69±0.06</td>
</tr>
<tr>
<td>20</td>
<td>-0.14±0.07*</td>
<td>0.90±0.06</td>
<td>-0.010±0.015</td>
<td>-0.15±0.08</td>
<td>0.90±0.06</td>
</tr>
<tr>
<td>30</td>
<td>0.70±0.10</td>
<td>1.02±0.09</td>
<td>-0.045±0.041</td>
<td>0.75±0.11</td>
<td>1.02±0.09</td>
</tr>
</tbody>
</table>

* From Khakoo and McConkey (1987).
Table 11. Parameters for Xe $6s\,^3\!\!\!\!\,^3P_1$ excitation at 30 eV impact.

<table>
<thead>
<tr>
<th>$\theta_{0,\text{deg}}$</th>
<th>$P_{1t}^M$</th>
<th>$P_{3t}^M$</th>
<th>$\rho_{00}$</th>
<th>$P_{t}$</th>
<th>$P_{3t}$'</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>0.42±0.06</td>
<td>0.75±0.09</td>
<td>-0.064±0.065</td>
<td>0.55±0.09</td>
<td>0.81±0.11</td>
</tr>
<tr>
<td>5</td>
<td>0.20±0.05</td>
<td>0.68±0.04</td>
<td>-0.030±0.021</td>
<td>0.27±0.07</td>
<td>0.71±0.04</td>
</tr>
<tr>
<td>10</td>
<td>-0.39±0.04</td>
<td>0.44±0.03</td>
<td>-0.013±0.014</td>
<td>-0.51±0.05</td>
<td>0.46±0.03</td>
</tr>
<tr>
<td>15</td>
<td>-0.59±0.04</td>
<td>0.16±0.04</td>
<td>0.023±0.021</td>
<td>-0.79±0.05</td>
<td>0.17±0.04</td>
</tr>
<tr>
<td>20</td>
<td>-0.45±0.03</td>
<td>0.21±0.05</td>
<td>0.060±0.025</td>
<td>-0.61±0.04</td>
<td>0.21±0.05</td>
</tr>
<tr>
<td>25</td>
<td>0.06±0.08</td>
<td>0.64±0.07</td>
<td>-0.012±0.033</td>
<td>0.08±0.11</td>
<td>0.64±0.07</td>
</tr>
<tr>
<td>30</td>
<td>0.48±0.08</td>
<td>0.89±0.10</td>
<td>-0.107±0.056</td>
<td>0.63±0.10</td>
<td>0.89±0.10</td>
</tr>
<tr>
<td>40</td>
<td>0.55±0.09</td>
<td>0.80±0.05</td>
<td>-0.049±0.033</td>
<td>0.73±0.12</td>
<td>0.80±0.05</td>
</tr>
<tr>
<td>50</td>
<td>0.00±0.08</td>
<td>0.57±0.10</td>
<td>0.011±0.049</td>
<td>0.00±0.10</td>
<td>0.57±0.10</td>
</tr>
</tbody>
</table>
Table 12. Parameters for Xe $6s \left| \frac{3}{2} \right| ^0 3P_1$ excitation at 50 eV impact.

<table>
<thead>
<tr>
<th>$0_\gamma$(deg)</th>
<th>$P^M_1$</th>
<th>$P^M_1$</th>
<th>$\rho_{00}$</th>
<th>$P_1$</th>
<th>$P_1^-$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>0.73±0.06</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0.11±0.08</td>
<td>0.63±0.04</td>
<td>-0.016±0.024</td>
<td>0.15±0.10</td>
<td>0.66±0.04</td>
</tr>
<tr>
<td>10</td>
<td>-0.43±0.03</td>
<td>0.38±0.04</td>
<td>-0.004±0.017</td>
<td>-0.57±0.04</td>
<td>0.40±0.04</td>
</tr>
<tr>
<td>15</td>
<td>-0.53±0.05</td>
<td>0.26±0.04</td>
<td>0.010±0.022</td>
<td>-0.71±0.07</td>
<td>0.27±0.04</td>
</tr>
<tr>
<td>20</td>
<td>-0.04±0.09</td>
<td>0.52±0.08</td>
<td>0.029±0.039</td>
<td>-0.06±0.12</td>
<td>0.52±0.08</td>
</tr>
<tr>
<td>30</td>
<td>0.12±0.13</td>
<td>0.81±0.12</td>
<td>-0.083±0.055</td>
<td>0.16±0.17</td>
<td>0.81±0.12</td>
</tr>
<tr>
<td>40</td>
<td></td>
<td>0.83±0.26</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 13. Parameters for Xe $6s \left| \frac{3}{2} \right| ^0 3P_1$ excitation at 80 eV impact.

<table>
<thead>
<tr>
<th>$0_\gamma$(deg)</th>
<th>$P^M_1$</th>
<th>$P^M_4$</th>
<th>$\rho_{00}$</th>
<th>$P_1$</th>
<th>$P_1^-$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>-0.31±0.04</td>
<td>0.37±0.06</td>
<td>0.012±0.028</td>
<td>-0.41±0.05</td>
<td>0.43±0.06</td>
</tr>
<tr>
<td>10</td>
<td>-0.58±0.06</td>
<td>0.16±0.05</td>
<td>0.023±0.032</td>
<td>-0.78±0.08</td>
<td>0.18±0.05</td>
</tr>
<tr>
<td>15</td>
<td>-0.19±0.14</td>
<td>0.53±0.13</td>
<td>-0.005±0.059</td>
<td>-0.25±0.19</td>
<td>0.54±0.13</td>
</tr>
<tr>
<td>20</td>
<td>0.29±0.25</td>
<td>0.84±0.23</td>
<td>-0.091±0.119</td>
<td>0.38±0.33</td>
<td>0.84±0.23</td>
</tr>
<tr>
<td>25</td>
<td></td>
<td>0.93±0.33</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 22. $P_1$ and $P_4$ parameters for 80 eV excitation of Ne. Open circles: present data; solid line: Bartschat and Madison (1987), DWBA.
Figure 23. $P^M_1$ and $P^M_4$ parameters for 80 eV excitation of Ar. Symbols as in Figure 22.
Figure 24. $P_1^M$ and $P_4^M$ parameters for 60 eV excitation of Kr $5s^1\{1/2\}^o$. Symbols as in Figure 22 except that the data of Bartschat and Madison have also been corrected for hyperfine interaction effects (solid line). The dashed line shows the DWBA data before this correction.
Figure 25. $P_4^M$ and $P_4^M$ parameters for 60 eV excitation of Kr $5s[3/2]^1$. Symbols as in Figure 24.
Figure 26. $P_1''$ and $P_4''$ parameters for 30 eV excitation of Xe $6s[3/2]^0$. Symbols as in Figure 24.
Figure 27. $P_1^M$ and $P_4^M$ parameters for 50 eV excitation of Xe $6s[3/2]^0$. Symbols as in Figure 24.
Figure 28. $P_1^M$ and $P_4^M$ parameters for 80 eV excitation of Xe $6s[3/2]^0$. Symbols as in Figure 24.
Figure 29. $\rho_{oo}$ parameter for 80 eV excitation of Ne. Circles: present data; solid line: Bartschat and Madison (1987), DWBA.
Figure 30. $\rho_{00}$ parameter for 80 eV excitation of Ar. Symbols as in Figure 29.
Figure 31. $\rho_0$ parameter for 60 eV impact excitation of the $5s\,^1{^1}\text{P}_1$ (top) and $5s\,^3{^1}\text{P}_1$ (bottom) states of Kr. Circles: present data; squares: from Danjo et al (1985); triangles: from King et al (1985); solid line: Bartschat and Madison (1987), DWBA; dashed line: Meneses et al (1985), FOMBT. All experimental data have been corrected to take account of hyperfine depolarisation.
Figure 32. $\rho_{\infty}$ parameter for electron impact excitation of Xe $6s\left(\frac{3}{2}\right)^0$ at 30 eV (top), 50 eV (middle), and 80 eV (bottom). Circles: present data; squares: from Nishimura et al (1986); triangles: from McGregor et al (1982); solid line: Bartschat and Madison (1987), DWBA. All experimental data have been corrected to take account of hyperfine depolarisation.
6.3 Coherence Results

This section presents new $P_2$ and $P_3$ results for excitation of the $6s[3/2]^0_1(1^P)$ state of Xe by 30 eV electrons. These results are then used with the already discussed $P_1$ results to derive measured coherence parameters. The data are listed in Table 14 and the measured Stokes parameters are shown in Figures 33 and 34. Also shown are the results of Nishimura et al (1986) and the theoretical DWBA data of Bartschat and Madison (1987). Included on the figures are two theoretical curves, the original data of Bartschat and Madison (1987) and also modified data in which hyperfine depolarisation has been taken into account. The dramatic influence of this effect in the $P_{2M}$ case is obvious. The agreement between the present data and theory is good over the entire angular range studied. Discrepancies at 5° and below are not unexpected given the rapidly varying nature of the $P_1$ and $P_2$ Stokes parameters in this angular region and the possible influence of finite interaction volume effects at these very small angles. The 10° datum point of Nishimura et al (1986) for $P_2$ seems to be clearly in error. It is noted that no other experimental circular polarisation ($P_3$) data is available.

The variation of the alignment angle, $\gamma$, of the excited state charge cloud with electron scattering angle is shown in Figure 35. The agreement between the measured data and theoretical results of Bartschat and Madison (1987) is excellent over the whole angular range. This ability of the DWBA to predict very good values of $\gamma$ has been recognised previously for this and other targets and impact energies (Andersen et al 1988, Murray et al 1990). Also shown on Figure 35 are the first Born approximation (FBA) theoretical predictions. This treatment uses plane waves in the incoming and outgoing channels and axial symmetry about $\Delta \vec{k} = \vec{k}_{in} - \vec{k}_{out}$ exists, meaning that since the ground state is a $1^S_0$ state, the excited state is a pure p-orbital in the direction of $\Delta \vec{k}$ with
\[ \ell^{kk} = 0 \]  \hspace{1cm} (6.3.1)

and

\[ P^{kk} = 1 \]  \hspace{1cm} (6.3.2)

The charge cloud alignment angle \( \gamma \) is given by

\[ \tan \gamma = \frac{\sin \theta}{\cos \theta \left[ \frac{E}{k_n} \right]^2} \]  \hspace{1cm} (6.3.3)

where \( E \) is the incident electron energy and \( E/L \) is the energy loss. Figure 35 indicates that the FBA has some validity in that it correctly indicates the direction in which the charge cloud starts to rotate.

The linear, \( P^l \), and total, \( P^r \), polarisations of the charge cloud are shown in Figure 36. In these figures the data have been corrected for hyperfine interaction effects and so they represent the situation which is applicable to the nascent charge cloud. It is seen that, again, very good agreement with the DWBA data is obtained except at angles \( \leq 15^\circ \) where, as mentioned above, experimental problems are expected to be most severe. Most significantly it is seen that, within experimental error, \( P^r \) is unity over the angular range studied (again omitting the data at \( 5^\circ \) and below). This result, coupled with the earlier result that \( \rho_{00} \) is zero over this angular range, establishes the complete coherence of the excitation for this range of electron scattering angles.
Table 14. Stokes parameter data for 30 eV excitation of Xe \( 6s[3d^7;^1P] \).

<table>
<thead>
<tr>
<th>( O_v )</th>
<th>( P^{M}_1 )</th>
<th>( P^{M}_2 )</th>
<th>( P^{M}_3 )</th>
<th>( Y^{''}_{(clos)} )</th>
<th>( P^*_{i} )</th>
<th>( P^* )</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>0.42±0.06</td>
<td>-0.32±0.06</td>
<td>-0.02±0.25</td>
<td>-19±3</td>
<td>0.70±0.08</td>
<td>0.70±0.09</td>
</tr>
<tr>
<td>5</td>
<td>0.20±0.05</td>
<td>-0.43±0.07</td>
<td>-0.34±0.17</td>
<td>-32±4</td>
<td>0.63±0.09</td>
<td>0.73±0.12</td>
</tr>
<tr>
<td>10</td>
<td>-0.39±0.04</td>
<td>-0.68±0.03</td>
<td>-0.36±0.04</td>
<td>-60±1</td>
<td>1.05±0.04</td>
<td>1.11±0.04</td>
</tr>
<tr>
<td>15</td>
<td>-0.59±0.04</td>
<td>-0.19±0.04</td>
<td>-0.51±0.06</td>
<td>-81±2</td>
<td>0.83±0.05</td>
<td>0.99±0.06</td>
</tr>
<tr>
<td>20</td>
<td>-0.45±0.03</td>
<td>0.42±0.10</td>
<td>-0.54±0.07</td>
<td>69±3</td>
<td>0.82±0.09</td>
<td>1.01±0.09</td>
</tr>
<tr>
<td>25</td>
<td>0.06±0.08</td>
<td>0.43±0.09</td>
<td>-0.66±0.14</td>
<td>41±5</td>
<td>0.58±0.11</td>
<td>0.92±0.14</td>
</tr>
<tr>
<td>30</td>
<td>0.48±0.08</td>
<td>0.22±0.08</td>
<td>13±4</td>
<td>0.71±0.10</td>
<td></td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>0.55±0.09</td>
<td>-0.13±0.08</td>
<td>-7±4</td>
<td>0.76±0.12</td>
<td></td>
<td></td>
</tr>
<tr>
<td>50</td>
<td>0.00±0.08</td>
<td>-0.48±0.12</td>
<td>-45±5</td>
<td>0.64±0.16</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* The angles are given in the first or fourth quadrant.
Figure 33. $P^*_2$ parameter for 30 eV excitation of Xe $6s\{3/2\}^0$. Circles: present data; dashed line: data of Bartschat and Madison (1987); solid line: data of Barstschat and Madison (1987) corrected for hyperfine depolarisation effects; crosses: data of Nishimura et al (1986).
Figure 34. $P_3$ parameter for 30 eV excitation of Xe $6s[3/2]^6$. Symbols as in Figure 33.
Figure 35. Variation of charge cloud alignment angle. Excitation is of the $6s[3/2]^0$ state of Xe at 30 eV. Symbols as in Figure 33 with the addition of the dotted line indicating the first Born approximation.
Figure 36. Variations of (a) linear and (b) total polarisations. Excitation is of the $6s[3/2]_1^0$ state of Xe. Symbols as in Figure 33.
Chapter VII

CONCLUSIONS

An experiment has been described in which electron-photon coincidence measurements were carried out using gaseous targets. These measurements yield the most basic possible information about the electron impact excitation process.

Models have been developed to investigate a number of effects which arise due to the finite volume of the interaction region and the finite acceptance angles of the electron and photon detectors. The effects have been found to be potentially particularly severe in the measurement of the $P_\sigma$ coherence parameter. Slight misalignment or defocussing of the electron beam through the nominal scattering center can have dramatic effects on the measured $P_\sigma$ particularly at small scattering angles and when $P_\sigma$ approaches -1. It is suggested that the use of Ne or Ar as target species can help identify the magnitude of any effect which might be present. In some cases placing the in-plane photon detector at $\pi / 4$ to the electron beam direction can be helpful.

Polarisation correlation measurements of the $P_\sigma$ and $P_\pi$ Stokes parameters have been presented for Ne, Ar, Kr, and Xe for incident energies in the range 30 - 80 eV and electron scattering angles up to 50°. After adequate account was taken of finite interaction volume and nuclear spin depolarising effects, good agreement was found with theoretical predictions. In particular, previous discrepancies between theory and experiment with regard to the charge cloud height parameter, $\rho_{oo}$, have been resolved.

When any internal atomic depolarising interaction was found to be occurring, so that the charge cloud acquired a "height", the effect on $P_\sigma$ measurements was much greater than any of the finite interaction region effects which were investigated.

A complete Stokes parameter analysis has been carried out for electron impact excitation of the $6s[3/2]^1$ resonance level of Xe at 30 eV energy. This work, which dealt with the part of the excitation which possessed positive reflection symmetry in
the scattering plane, showed that the DWBA behaves very well at least for scattering angles up to 50°. This, coupled with the $\rho_{00}$ results, allows the conclusion that the excitation is essentially completely coherent for the range of experimental parameters investigated. Complete hyperfine depolarisation was observed to occur before the decay of the excited state.

Future directions for this experiment involve installation of LiF filters on the photon detectors. Such a filter, with its very sharp cutoff at 105 nm, will allow isolation of the $Ar + s[3/2]_0^0$ state which, according to theoretical calculations, displays interesting $\rho_{00}$ characteristics at low impact energies and at scattering angles within the range of the present studies. For this state $\rho_{00}$ departs significantly from zero and the total polarisation $P$ drops substantially from unity. Such measurements should provide a sensitive test of the ability of theories such as DWBA to describe the excitation process when significant spin flip is occurring. A further development will be the installation of an indirectly heated cathode in place of the existing tungsten filament. This should greatly reduce the energy spread of the electron beam and may lead to a resolution such that states like $Ar + s'[1/2]_0^0$ and $+s[3/2]_0^0$ may be separated.
REFERENCES


Fano U and Macek J H 1973 Rev. Mod. Phys. 45 553-573


Harting E and Read F H 1976 Electrostatic Lenses Elsevier: Amsterdam


O'Neill E L 1963 Introduction to Statistical Optics Addison-Wesley: Reading, Mass. 133-156


Slevin J 1984 Rep. Prog. Phys. 47 461-512


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Jay Corr was born in Windsor, Ontario on Sept. 15, 1957. He graduated from Walkerville C. I. in 1975. His next four years were spent as a professional alpine skier based out of Whistler, B.C., followed by a year of managing a large drug mart-department store in Calgary, Alberta.

In 1981 Jay enrolled at the University of Windsor and financed his way through undergraduate school by working part-time at one of the local automobile factories. He graduated, in 1985, with an honours B.Sc. with a major in physics. In his second year of graduate school Jay received a University of Windsor tuition scholarship and then obtained his M.Sc. in physics in 1987. NSERC awarded Jay a scholarship for his first two years of doctoral studies and he is currently a doctoral candidate hoping to graduate in Fall 1990.

During Jay's third year of undergraduate school he was married and in 1987 he and his wife had their first child.
PUBLICATIONS


CONFERENCE PRESENTATIONS


GEC 1988 Corr J J, Plessis P and McConkey J W Electron-Photon Coincidence Studies of Heavy Rare Gas Excitation by Electron Impact


