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Electron Collisions with Metastable Helium*

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Abstract

Measurements and theoretical studies of electron scattering from the metastable $2^{1,3}$S excited states of helium are briefly reviewed. An apparatus developed for the study of low energy differential scattering from metastable excited states is discussed in detail and the first results of angular distributions for electrons superelastically scattered from the $2^{1,3}$S states at incident energies of 5, 10 and 30 eV are presented.

1. Introduction

Electron collisions with excited atoms, either those in optically accessible or metastable excited states, are important in a range of situations including low temperature plasmas, discharge-based devices such as lamps and lasers and in stellar and planetary atmospheres. Because of the long natural lifetimes of the metastable states (e.g. ~8000 s for the 2^3S state of He) and their large internal energy and scattering cross sections, they can be present in relatively small concentrations and still have a significant effect on the macroscopic behaviour of the system. An understanding of both the nature and magnitude of these collision processes is fundamental to a complete understanding of such systems and devices. Excited state collision processes also provide an interesting theoretical challenge. The atomic structure and subsequent collisional properties of these states are typically quite different to those of the parent ground state and, as such, the approximations that may be appropriate for a description of ground state collision processes may need to be revised.

Despite the above, the body of experimental information on metastable-excited state collisions, particularly those in which absolute values have been obtained for scattering cross sections, is rather small. For the experimentalist the difficulty in performing electron scattering experiments from metastable species arises principally from the low target populations which are achievable with conventional excited state sources. For example, in a crossed beam experiment, metastable excited state target densities of the order of the 10^8 cm^{-3} are possible, but these are small compared to beam densities for typical ground state target experiments which may be in excess of 10^{12} cm^{-3}. Such excited state experiments,

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with their subsequent low signal levels, are often further hampered by large background contributions due to charged particles and UV photons that arise from the processes used to produce the excited states. Also, Penning ionisation of impurities (e.g. N_2) in the gas beam by the metastable atoms can futher result in large numbers of low energy electrons which present a real problem for inelastic scattering measurements (see for example Müller-Fiedler*et al.* 1984). These Penning processes, which occur with large cross sections ($\approx$100 Å^2), also contribute to limiting the ultimate number density achievable in such an excited state beam. As a result of these and many other limitations, experimental work in this field lags well behind theory.

There have been several excellent reviews of electron–excited-atom scattering in recent years (Lin and Anderson 1991; Trajmar and Nickel 1992) and it is not our intention to duplicate those here. Rather we shall give a short, specific yet by no means complete, discussion of experiments and theoretical calculations on scattering from the metastable states of helium, before discussing an apparatus which we have developed for low energy electron scattering from rare gas metastable atoms and the application of this apparatus in the first differential (in angle) studies of superelastic scattering from the $2^{1,3}$S states of helium. A brief report of some of this work has appeared in Jacka *et al.* (1995).

2. Brief Review of the Field

The progress in experimental work in this field roughly parallels that in the development of the technology of metastable atom sources. Grand total cross sections (GTCS) for electron scattering from 2^1S and 2^3S helium atoms have been measured by Neynaber *et al.* (1964) and Wilson and Williams (1976) at incident energies between 0·5 and 10 eV. Both of these investigations employed a hot-cathode, low-voltage discharge as the source of metastable atoms and both measured the attenuation of the metastable beam by a transverse beam of electrons. These experiments revealed extremely large (400–900 Å^2) GTCS for both metastable species at energies of a few eV. Such cross sections are at least two orders of magnitude larger than the corresponding measurements for scattering from the ground state of helium.

There is a fairly substantial body of experimental measurements, principally by Lin, Anderson and co-workers, of total cross sections for electron excitation out of the metastable levels to discrete, higher excited states. These experiments (see for example Rall *et al.* 1989; Lin and Anderson 1991) used a hollow cathode discharge to produce a beam of $2^{1,3}$S helium atoms which is then crossed by a low energy electron beam. They measured absolute, apparent excitation cross sections for a large number of these higher excited states by observing the yield of photons emitted when these states, excited from the 2^3S level by electron impact, decay to a lower lying level. These measurements are only reliable for relatively low incident electron energies (<22·7 eV). Higher energies result in excitation of the substantial fraction of ground state atoms in the atomic beam, from which the resultant radiation swamps the metastable level measurements. When the incident electron energy is greater than the second excitation energy above the metastable level more than one photon can be emitted in a cascade, so extracting the cross sections requires a knowledge of the branching ratios of the photon decay processes. Typical total cross sections for excitation to the

3^3S, 3^3P, 3^3D levels lie in the range 5–10 Å^2.

To our knowledge, the work of Müller-Fiedler *et al.* (1984) represents the only previous experimental investigation of differential (in angle) electron scattering from He (2^3S). This work involved the measurement of angular distributions for excitation of a large range of final states (2^3P, 3^3S, 3^3P, 3^3D, and to the sum of the 4^3L states) by use of an energy resolved electron beam. The excited atomic beam was produced in a cold-cathode DC discharge, the incident electron energies were 15, 20 and 30 eV, and the range of scattered electron angles varied between 10° and 40°. Absolute magnitudes were assigned from estimates of the ratio of excited state to ground state populations in the beam and by comparison with the ground state elastic cross section. The resultant differential cross sections were all strongly enhanced in the forward direction, particularly that for the 2^3S–2^3P transition which, at an incident energy of 15 eV, attains a value of $\approx$500 Å^2 sr^{-1} at a scattering angle of 10° and drops by more than three orders of magnitude by 40°.

A number of theoretical approaches have been used to study elastic and inelastic electron scattering from metastable helium. These include multichannel eikonal theory (Flannery and McCann 1975; Mansky 1990), distorted wave approximations (Mathur *et al.* 1987), first order many body theory (França and da Paixão 1994; Cartwright and Csanak 1995) and convergent close coupling calculations (Bray and Fursa 1995). One interesting aspect of all of these theoretical models is the prediction of a sharp dip at forward angles in the 2^3S–3^3P differential cross section which was not observed by Müller-Fiedler *et al.* (1984). Calculated cross sections for superelastic scattering from the metastable levels can be readily obtained, via detailed balance theory, from the large number of inelastic scattering calculations for the reverse processes that exist in the literature.

To a large extent the nature of the cross sections for electron scattering from rare gas metastable atoms, and helium in particular, can be readily understood by a comparison with both the structure and electron scattering cross sections of their nearest alkali neighbours. With the obvious exception of the structure of the core and the coupling scheme appropriate for the description of fine-structure levels, He(2^3S) is in many respects similar to lithium. For example their first excited resonance levels occur at very low energies (1·14 and 1·84 eV respectively) and they have similar ionisation potentials (4·76 and 5·39 eV). In particular their dipole polarisabilities, which are a useful measure of the magnitude of their low energy electron scattering cross sections, are also very large [He(2^3S)—301 a.u., He(2^1S)—728 a.u. (Crosby and Zorn 1977), Li(2^2S)—164 a.u. (Miller and Bederson 1977)]. Dipole polarisabilities are larger in the alkali metals than for all other ground state systems, and this is reflected in their large electron scattering cross sections. That the dipole polarisabilities of metastable helium He ($2^{1,3}S$) are substantially larger again helps provide some intuitive insight into the reactivity of these loosely bound metastable species.

3. Experimental Apparatus

The experiments described here have been carried out with a crossed electron–excited-atom beam apparatus which is shown schematically in Fig. 1. The apparatus consists of two differentially pumped vacuum chambers, one of which, the source chamber, houses the atomic beam source and the other, the scattering

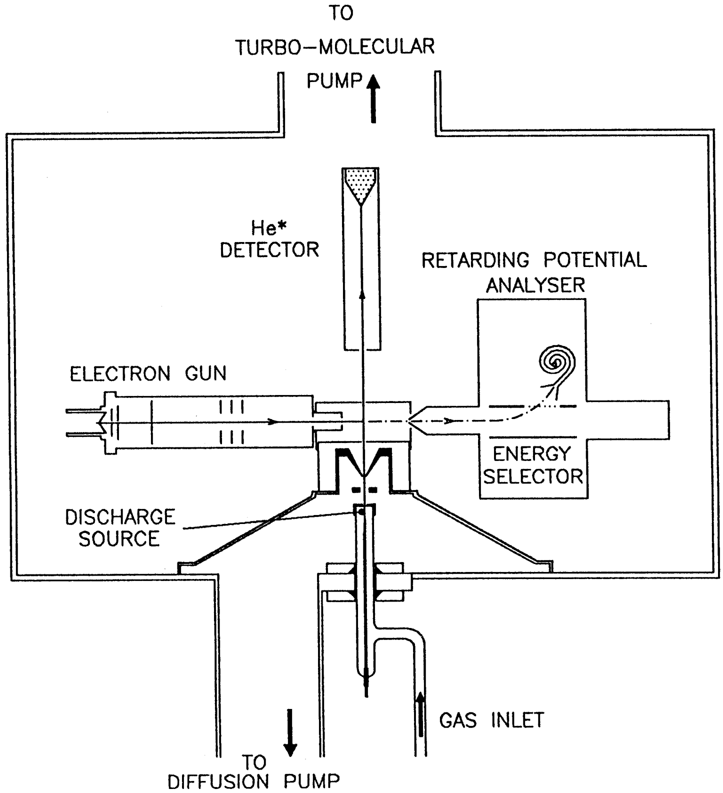

Fig. 1. Schematic diagram of the crossed beam apparatus showing the beam source and scattering chambers and elements of the discharge source and electron spectrometer.

chamber, contains the electron spectrometer used for the scattering measurements. The source chamber is pumped with a 1000 $\mathrm{l\,s^{-1}}$ diffusion pump, although physical restrictions around the source exit provide the ultimate limitation on the pumping speed in this area. The scattering chamber is pumped by a 550 $\mathrm{l\,s^{-1}}$ turbomolecular pump. Base pressures in these chambers are 1×10^{-7} and 1×10^{-8} mbar respectively. When operational, the differential pumping between the source and scattering chamber enables the background pressure in the scattering chamber to be maintained at $\sim7\times10^{-7}$ mbar whilst that in the source chamber is as high as 1×10^{-4} mbar. Low background pressures are particularly important in order to reduce collisional quenching of the metastable atoms and also to minimise the production of UV photons in the electron gun and scattering region. All materials used in the construction of the apparatus are non-magnetic and the scattering chamber is lined with a sheath of high permeability magnetic shielding. In addition a pair of Helmholtz coils are used to counteract the major (vertical) component of the Earth's magnetic field.

For the present scattering experiments the electron beam is generated by a simple electron gun consisting of a triode stage, to extract electrons from a thoriated tungsten filament, followed by a three element aperture lens to focus the electrons and define their final energy. The energy range is from 5 to 30 eV with electron beam currents of ~5 to 60 μA respectively. Because of the large current and corresponding space charge effects, and the lack of a monochromation stage in the gun, the energy spread in the electron beam from the gun is estimated to be about 700 meV. The electron beam crosses the atomic beam at 90° in the centre of a small, mesh-enclosed interaction region. The electron beam current is monitored with a Faraday cup which faces the electron source from the opposite side of the scattering region. Electrons which are scattered by the atomic beam are detected by an analyser which can be rotated in a plane containing the electron beam and perpendicular to the atomic beam (the scattering plane). In the present experiment we are interested in those electrons which have been superelastically scattered from the metastable helium beam, resulting in the scattered electron carrying off the excitation energy of 19·81 eV in the case of the 2^3S state. The superelastically scattered electrons enter the analyser and are deflected through ~60° by a transverse linear electric field, to separate them from background UV photons, before passing a single grid retarding potential analyser and being accelerated onto an electron multiplier. The overall energy resolution of the analyser and electron beam is ~1 V which is more than adequate to resolve the superelastically scattered electrons from those that have been elastically or inelastically scattered.

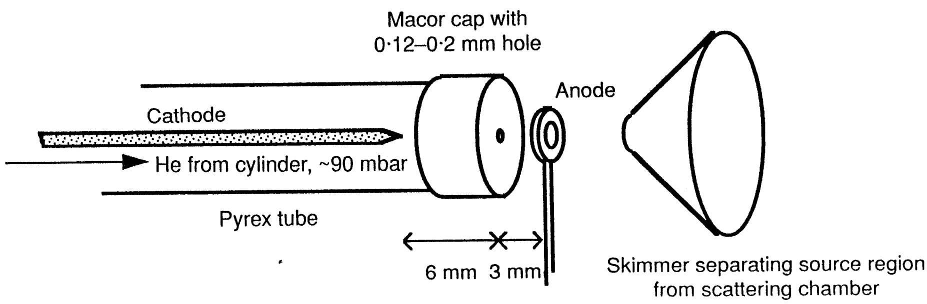

Fig. 2. The cold-cathode metastable atom source.

The metastable atoms are produced in a cold-cathode DC discharge (Fig. 2). This consists of a 10 mm diameter pyrex tube, through which the helium is introduced, which contains a tungsten needle cathode and which has a macor (machine ceramic) end cap with a 120 mm diameter central hole. The discharge is struck between the tungsten needle cathode (−700 V), through the hole in the macor cap to a tungsten ring anode (+200 V) located about 3 mm downstream of the cap. The flow of ultra-high purity helium (99·995%) to the discharge is controlled by an automatic flow controller which maintains a driving pressure in the pyrex tube of between 50 and 100 mbar, depending on the state of wear of the hole in the macor cap. The discharge draws a current of ~10 mA via a 40 KΩ load resistor. The high pressure region between the needle and the cap is required to maintain the stability of the discharge and most of the metastable atoms produced there are effectively quenched by subsequent collisions in the expansion through the nozzle. The substantial part of the useable metastable atom flux is produced in the low pressure region between the cap and anode. A 1 mm diameter skimmer is located a further 10 mm downstream from the anode and the helium beam passes through this into the scattering chamber. Differential pumping is applied across the skimmer.

4. Diagnostic Techniques for Metastable Rare Gas Sources

There have been quite a number of different metastable rare gas sources designed in the last thirty years or so for a variety of purposes. These include studies of atom–atom interactions such as Penning ionisation (Müller *et al.* 1991), surface interactions (e.g. Riddle *et al.* 1981), electron–atom interactions such as the work described here and photon–atom interactions for cooling and trapping experiments (e.g. Bardou *et al.* 1992). There may be different requirements for each purpose. For example, for electron scattering experiments, the primary requirement is for as many atoms as possible to be in the interaction volume, which must be free from magnetic fields. For laser cooling the velocity distribution and number density (in perhaps a very small volume) may be important. As well as the number density and flux of metastable atoms coming from a source, their transverse and longitudinal velocity distributions and the presence of photons, ions and electrons may also be important. It is the purpose of this section to describe the various techniques which have been developed to determine the output (metastable species, flux, velocity distributions, etc.) from metastable atom sources.

The large excitation energy of the metastable excited states of the rare gases means that they can be detected rather simply by collecting the secondary electron that is emitted when they strike a surface, for example an electron multiplier or bare metal. The efficiency of this process has been determined for a number of surfaces (e.g. Dunning *et al.* 1975) and, although it is very much dependent on the condition of the surface, it is generally larger than 60% for helium. Unfortunately UV photons, electrons and ions are also detected in a similar fashion, although the efficiency of detection for the photons is generally less than 10%. In beams produced by discharge-based sources, UV photons and charged particles can outnumber metastable atoms by many orders of magnitude, and the relative populations of different species in the beam can be very sensitive to subtle changes in the external parameters governing the discharge behaviour. Electrons and ions can be removed using electrostatic deflectors, although in a discharge-based source the use of the cathode as the upstream electrode provides no guarantee of complete removal of positive ions from the effusing atomic beam as they can still be formed in large quantities in the afterglow. For example, in our source it is possible to measure microamps of positive ion current if there is inadequate electrostatic deflection, compared to (typically) tens of nanoamps of true metastable yield. Diagnostically, UV photons in the beam can be separated from the metastable atoms by nature of their much greater velocity. This can be simply done by chopping the beam with a rotating slotted wheel and, with the aid of an electron multiplier placed downstream, a time-of flight spectrum can be obtained which contains photon and metastable atom contributions which are well separated in time. Such a measurement also gives information about the longitudinal velocity distribution of the atoms which, for discharge-based sources, can be affected by the type of expansion in the nozzle and collisions within the beam (Hardy and Sheldon 1981). The transverse velocity distribution can be obtained from measurements of the angular distribution.

Alternative diagnostic techniques utilise optical transitions from the metastable level(s). When the metastable density is high, such as in or near the output of a discharge, the density can be measured by laser photoabsorption at a transition frequency (e.g. Kane and Dunn 1983). In some systems, for example neon, it is possible to quench the entire metastable population by optically exciting all of the metastables to levels which decay preferentially to the ground state. The concurrent drop in metastable signal, such as a drop in the secondary emission current from a surface, can then be used as an accurate measure of the total metastable contribution in the beam (e.g. Kawanaka *et al.* 1993). A third method is laser induced fluorescence which has been successfully used to measure the number density of a metastable helium beam (e.g. Rall *et al.* 1989). The velocity distribution can be determined with all of these techniques by making use of the Doppler effect.

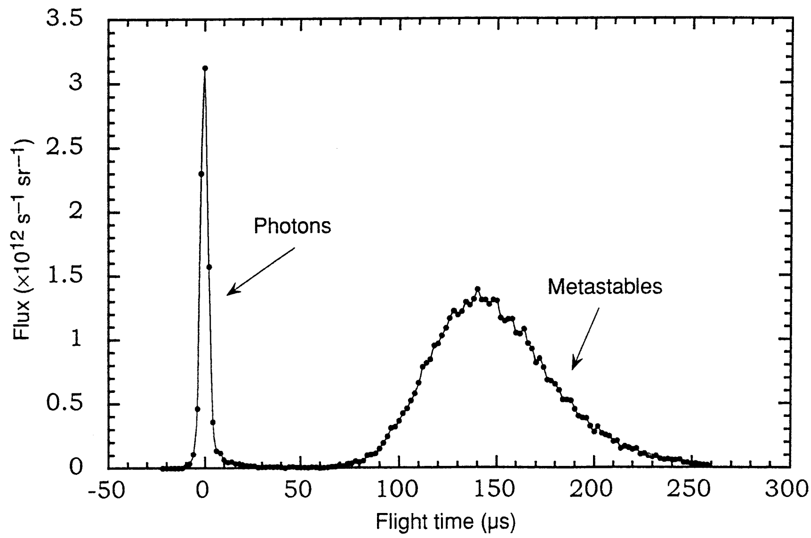

Fig. 3. A time-of-flight spectrum obtained by chopping the atomic beam, illustrating the prompt photon peak and the broad, near-Maxwellian peak due to metastable atoms.

In the present work, optical techniques could not be used to probe the population of the 2^3S state as the number densities are too low for photoabsorption measurements and it is not possible to efficiently quench this state by single photon absorption. As a result we have used the time-of-flight (TOF) technique, in combination with continuous monitoring of the secondary emission current ejected from a stainless steel beam trap, to fully characterise the metastable source. The atomic beam was chopped by a 6 cm diameter disc rotating at about 5000 rpm with two diametrically opposed 50 μm wide slots machined in its periphery. The size of the atomic beam was reduced for these measurements by inserting a 1 mm diameter aperture before the chopper. An electron multiplier placed 30 cm downstream from the chopper was used to detect metastable atoms and UV photons in the beam. To reduce the number of metastable atoms that reached the detector, and avoid saturation of it by high instantaneous count rates, it was placed behind another aperture which was 125 μm in diameter. A typical TOF spectrum obtained with this arrangement is shown in Fig. 3. It clearly shows the prompt peak due to detection of photons, the width of which indicates the timing resolution of about 6 μs, and the broad roughly Maxwellian peak due to the metastable atoms. The analysis of such spectra, together with the beam trap measurements (assuming a 100% detector efficiency for the metastable atoms), indicates that the typical metastable flux is 2×10^{13} atoms $\mathrm{sr^{-1}\,s^{-1}}$ with a mean velocity of 1800 $\mathrm{m\,s^{-1}}$. This corresponds to a number density in the interaction volume of about 4×10^{7} $\mathrm{cm^{-3}}$.

Whilst it was not practical to use optical methods for the measurement of the dominant 2^3S component of the beam it is possible to efficiently quench the 2^1S state and thus determine the ratio of ^{3}S to ^{1}S metastable atoms in the beam. To achieve this a high pressure, spiral-shaped helium discharge lamp was placed around the atomic beam as it entered the scattering chamber. The 2^1S atoms are removed by optically pumping to higher lying n^1P levels which have a strong preference for decay to the ground state. An associated measurement of the change in the ejected electron current from the metastable beam trap indicates the ratio of 2^1S to 2^3S atoms in the beam was 1 : 10.

5. Superelastic Scattering Results

As the first step in a programme of electron scattering from the metastable states of helium, differential cross sections for the de-excitation of metastable helium by electron impact (superelastic scattering) have been measured.

The count rate of superelastically scattered electrons was typically 0·1–0·3 Hz whilst the background levels were 1–2 Hz. When the analyser was set to detect scattered electrons at or below 40 eV (incident electron energy 20 eV) the background was predominantly due to UV photons from the discharge. When the incident electron energy was above 20 eV the background was mainly caused by UV photons produced by excitation of the background gas in the scattering region. In both cases there were further contributions from UV photons produced by electron impact excitation of the background gas in the higher energy electrostatic focussing regions within the electron gun. These relatively large background contributions were determined by modulating both the electron beam and RPA detector as well as making measurements with the gas discharge turned off.

To determine a differential cross section absolutely, in addition to the rate of scattered electrons, one must accurately know the number of electrons and target atoms in the interaction volume, the efficiency of the detector and its acceptance solid angle, or else be able to apply a normalisation process such as the relative flow technique. In the present study neither was possible and, as a result, only the shape of the differential cross section could be determined. Furthermore, it was also necessary to correct for systematic angular dependent effects, the largest of which was the variation in the overlap between the analyser viewing angle and the scattering region. This was determined by measuring the shape of the differential cross section for electrons elastically scattered from the ground state helium beam and normalising to the known shape (Nesbet 1979; Brunger *et al.* 1992).

Because of physical constraints it was not possible to quench the 2^1S state in the manner outlined above whilst the scattering experiments were being performed and as a result the atomic beam contains both singlet and triplet states in the ratio 1 : 10. The present results are therefore compared to a number of theories for which the singlet and triplet components have been added in the same ratio.

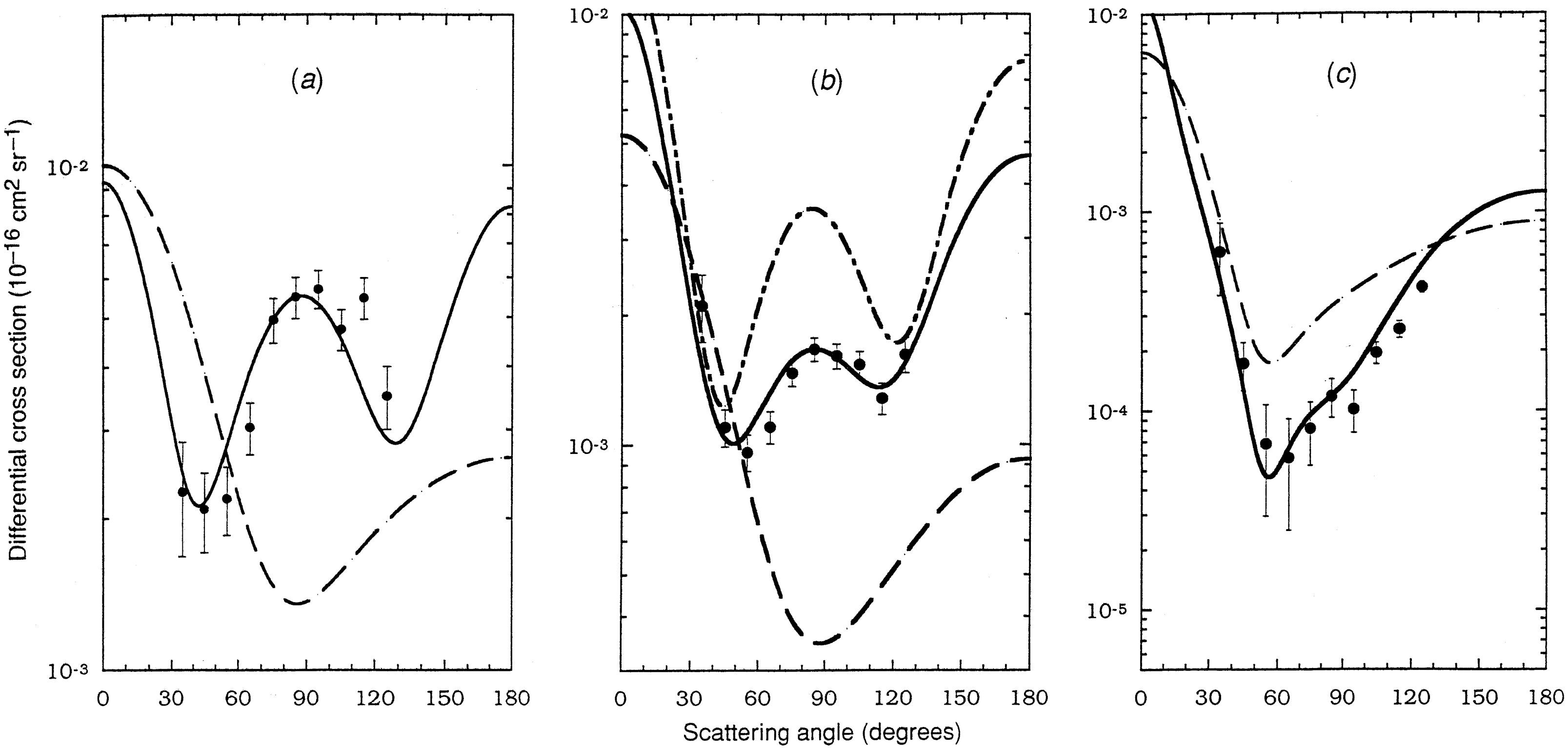

Fig. 4. Differential cross sections for superelastic scattering from metastable helium at incident electron energies of (*a*) 5 eV, (*b*) 10 eV and (*c*) 30 eV. Shown are the present results (experimental points), the convergent close coupling calculation (solid curves), the first order many body theory calculation (dashed curves) and the 29-state *R*-matrix calculation (dot–dash curve).

The measured differential cross sections are presented in Figs 4*a*, 4*b* and 4*c* for incident energies of 5, 10 and 30 eV respectively. They have been normalised in magnitude to the convergent close coupling (CCC) calculation of Bray *et al.* (1994) at a scattering angle of 85°. Cross sections from a 29-state *R*-matrix calculation (Fon *et al.* 1994) and a first order many body theory (FOMBT) calculation (Trajmar *et al.* 1992) are also included for comparison. They are presented as superelastic cross sections, having been derived from the (reverse) excitation cross sections by applying the principal of detailed balance. This relates the excitation $\sigma_{\rm e}$ and de-excitation $\sigma_{\rm d}$ cross sections via the energy of the incident electron $E_{\rm i}$, the excitation energy of the metastable level E and the ratio of degeneracies of the final and initial state g as

$$\sigma_{\rm d} = \sigma_{\rm e}\,\frac{E + E_{\rm i}}{gE_{\rm i}}\,.$$

Despite the fact that the CCC calculation is used as the normalisation it is clear that the present results are in best agreement with it at each of the energies shown. This is particularly true at the lowest energy of 5 eV (Fig. 4*a*) where there is little resemblance between the present angular distribution and that predicted by the FOMBT. The essential difference between the *R*-matrix and CCC calculations lies in the number of discrete coupled channels that are included (29 and 65 respectively) and also that continuum states are included in the CCC calculation. For the energies studied here the greatest difference between the two calculations occurs at 10 eV (Fig. 4*b*) and here the present results are clearly in better agreement with the CCC calculation. It is also apparent that as the energy is increased to 30 eV (Fig. 4*c*) the level of agreement with the FOMBT calculation improves as one might intuitively expect. The overall level of agreement between the present measurements and each of the theoretical curves is also consistent with that shown between several measurements of the reverse (1^1S–$2^{1,3}$S) excitation processes (Brunger *et al.* 1990; Trajmar *et al.* 1992) and each of these theories (Bray *et al.* 1994; Fon *et al.* 1994).

6. Conclusions and Future Directions

Low energy electron scattering from metastable excited atoms continues to attract the attention of the atomic physics community. With such experiments in mind, recent advances in the design of sources of metastable rare gas species mean that the current dearth of experimental results will soon be addressed. To this end we have constructed an apparatus to measure differential cross sections for a wide range of electron–metastable rare gas collision processes and have used it, in the first instance, to successfully measure differential cross sections for superelastic scattering from $2^{1,3}$S helium. As well as being the first such experiment of its kind, a number of useful techniques have been developed and the viability of the apparatus for further studies has been demonstrated. These measurements are in excellent agreement with the converged close coupling calculation of Bray *et al.* (1994).

The theoretical calculations of Bray and Fursa (1995), Cartwright and Csanak (1995), França and da Paixão (1993), Mathur *et al.* (1987) and Flannery and McCann (1975) all predict a sharp structure (dip) in the differential cross section for the 2^3S–3^3P excitation at forward angles, which was not observed by Müller-Fiedler *et al.* (1984), and which warrants further investigation. There are also some consistent differences between these data and theory concerning the absolute magnitude of the differential cross sections which, considering the difficulty of the experiments, are perhaps not surprising but which also warrant further attention. Furthermore, at energies lower than 15 eV there is a complete lack of experimental data on differential cross sections for inelastic scattering from the metastable levels. At energies below 5 eV one also expects to see strong effects due to the excitation of intermediate negative ion resonances. To examine these processes we have constructed a high resolution hemispherical electron spectrometer with a position sensitive detector. This will greatly assist in the measurement of excitation processes characterised, amongst other things, by extremely low count rates.

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