Journal of Physics B: Atomic, Molecular and Optical Physics

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To cite this article: S Wang *et al* 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 329

J. Phys. B: At. Mol. Opt. Phys. 27 (1994) 329–339. Printed in the UK

Coherent excitation of Ar $4s'[\frac{1}{2}]_1^0$ and $4s[\frac{3}{2}]_1^0$ by low energy electrons

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Received 23 April 1993, in final form 30 September 1993

Abstract. Electron–photon coincidence measurements are reported of the excitation of the $4s'[\frac{1}{2}]_1^0$ state of Ar at incident energies of 20 and 30 eV and scattering angles up to 80°. All values of the ρ_{00} parameter obtained are very small and not distinguishable from zero within experimental uncertainty. The level of agreement with available perturbative theories is quite good but significant discrepancies are noted at some angles particularly at the lower impact energy. Previous measurements from this laboratory of the $4s[\frac{3}{2}]_1^0$ state are reanalyzed in terms of the ratio of the exchange to direct excitation cross sections.

1. Introduction

Recent activity in the field of electron–photon coincidence experiments involving the heavy rare gases has been focused on obtaining data at incident energies approaching the threshold region where available theories might be suspect. These theories are all first-order perturbative in nature and represent various distorted wave approximations. They have been shown to provide very reasonable results in the higher energy region, i.e. at energies greater than about five times the threshold energy (see reviews by Andersen *et al* 1988, Becker *et al* 1992).

In this paper we present measurements of the P_1 and P_4 parameters for the $4s'[\frac{1}{2}]_1^0$ state of argon at electron impact energies of 20 and 30 eV. These measurements complement results for the $4s[\frac{3}{2}]_1^0$ state reported by Corr *et al* (1992). The purpose of both the present work and the previously reported results was to search for non-zero values of the ρ_{00} parameter and to stimulate further theoretical analysis of this difficult but important topic.

The most commonly used set of parameters to present and discuss results of coherence parameter measurements and calculations is the natural, frame-independent set introduced by Andersen *et al* (1986, 1988). These parameters, γ, $L_\perp$, P_{lin} and ρ_{00}, plus a differential cross section, give the most transparent description of the excitation process in terms of the excited state charge cloud (or oscillator density) characteristics. These define the charge cloud in and perpendicular to the scattering plane and the

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angular momentum transfer in the collision. They are related in a particular simple way to measured polarization correlation (Stokes) parameters as discussed below.

Since we are interested in charge cloud determinations we concentrate on the P_1 and P_4 linear polarization parameters and the ρ_{00} parameter. The ρ_{00} parameter gives the relative probability for spin-flip perpendicular to the scattering plane and, in terms of charge cloud characteristics, it gives the relative height of the charge cloud. For the noble gases ρ_{00} is related to the measured Stokes parameters by the following equation (Andersen *et al* 1986, 1988):

$$\rho_{00}=\frac{(1+P_1)(1-P_4)}{4-(1-P_1)(1-P_4)}$$

where P_1 and P_4 monitor the number of coincidences with photons polarized parallel to the incident electron beam minus those polarized perpendicular to this beam normalized to the total number of coincidence events, as detected by photon polarization analysers placed above the scattering plane (P_1) and in the scattering plane (P_4) respectively. In a polarization correlation experiment both in-plane and out-of-plane polarization measurements are required. Similarly if angular correlation experiments are carried out, the radiation pattern must be probed in three dimensions.

Clearly before any evaluation of ρ_{00} is made the measured Stokes parameters must be corrected for any experimental or other depolarizing effects. Van der Burgt *et al* (1991) have given an extensive discussion of the effects due to a finite interaction volume and due to hyperfine interaction. For our experimental set-up, finite volume effects were shown to be negligible for all scattering angles considered in the present work. Also, by using Ar as the target gas nuclear spin effects were eliminated.

2. Experimental details

The experimental procedures involved in the measurements of coherence parameters have been reported earlier (Corr *et al* 1991, 1992), and the reader is referred to these reports concerning the experimental apparatus and measuring techniques used.

The fine structure splitting of 0.19 eV between the two 4s states in Ar is too small to be resolved except by the use of an electron monochromator. Another approach is to use a LiF filter (which has a temperature dependent cut-off around 105 nm) to transmit the 106.7 nm photon emitted by the $4s[\frac{3}{2}]^0_1$ state and block the 104.8 nm photon from the $4s'[\frac{1}{2}]^0_1$ state. This approach was used to measure the coherence parameters reported by Corr *et al* (1992). The measurements reported here were performed without a LiF filter in front of the photon detectors, so that the measured polarizations are primarily due to the $4s'[\frac{1}{2}]^0_1$ state with about a 30% contribution from the $4s[\frac{3}{2}]^0_1$ state. The measurements can be corrected for this contribution based on the ratio of the differential cross sections.

$$\alpha(\theta,E)=\frac{d\sigma}{d\Omega}[\tfrac{3}{2}]\Big/\frac{d\sigma}{d\Omega}[\tfrac{1}{2}]. \tag{1}$$

It follows that the measured polarization, P_M, can be expressed as:

$$P_M=\frac{1}{1+\alpha}P[\tfrac{1}{2}]+\frac{\alpha}{1+\alpha}P[\tfrac{3}{2}] \tag{2}$$

from which one obtains:

$$P[\tfrac{1}{2}]=(1+\alpha)P_{\rm M}-\alpha P[\tfrac{3}{2}]. \qquad (3)$$

The coherence parameters for the $4s'[\frac{1}{2}]_1^0$ state are obtained from the measured parameters $P_{1,{\rm M}}$ and $P_{4,{\rm M}}$, and the $P_1[\frac{3}{2}]$ and $P_4[\frac{3}{2}]$ parameters reported by Corr *et al* (1992), using differential cross sections measured by Chutjian and Cartwright (1981).

Corr *et al* (1992) did not measure $P_1[\frac{3}{2}]$ and $P_4[\frac{3}{2}]$ data for 15°, 20° and 25° scattering angles at 20 eV. In order to correct our experimental data, we have used newly measured values of 0.05 ± 0.27, -0.57 ± 0.21 and -0.92 ± 0.25 for $P_1[\frac{3}{2}]$ at 20 eV and 15 °, 20 °, 25 ° scattering angles, respectively. Attempts to measure $P_4[\frac{3}{2}]$ at these angles have failed due to the low intensity of the photon signal: at these angles P_1 approaches -1 (see van der Burgt *et al* 1991). Instead we have used theoretical DWBA calculations by Bartschat and Madison (1987). The error bars in these theoretical values have arbitrarily been chosen to be 0.30 to reflect reasonable uncertainties in the theoretical calculations.

The errors in the $P_1[\frac{1}{2}]$ and $P_4[\frac{1}{2}]$ parameters are evaluated taking into account the experimental errors in $P_{1,{\rm M}}$ and $P_{4,{\rm M}}$, in $P_1[\frac{3}{2}]$ and $P_4[\frac{3}{2}]$, and in α. The error in α is taken to be 10%, which is the error in the deconvolution of each feature in the energy loss spectra measured by Chutjian and Cartwright (1981). They take into account other errors in obtaining absolute differential cross sections, but these errors do not apply here because we are dealing with the ratio of cross sections. However, we have added in quadrature a 10% interpolation error for those angles where Chutjian and Cartwright (1987) do not list any cross sections.

Parameters that are important in obtaining reliable values for the coherence parameters are the polarization efficiencies of the reflection polarizers and the scattering angle. Before and after every coincidence measurement the non-coincidence polarizations are measured with both photon detectors in order that corrections due to small fluctuations in the polarization efficiencies can be made. On these occasions the alignment of the electron beam is checked by monitoring the minimum in the current on the analyser entrance aperture in a narrow range around 0°. We estimate the uncertainty in the aligning of the electron beam to be $\pm0.3°$. The uncertainty of setting the analyser at a particular scattering angle is $\pm0.3°$, resulting in an estimated uncertainty in the scattering angle of less than 0.6°.

The alignment of the electron beam and the correct determination of the electron scattering angle are of some relevance because of disagreements with other experiments—see the discussion in section 4. Two checks on the alignment were made. As a first check we measured the $P_{1,{\rm M}}$ parameter at 30 eV at $-10°$ to be 0.164 ± 0.051. This compares with the measurement at $+10°$ of -0.100 ± 0.036. Given that the $P_{1,{\rm M}}$ parameter is a symmetric function with scattering angle around zero degrees and has the value 1 at 0°, these measurements indicate a probable deviation in the scattering angle of $+0.94\pm0.23$ deg. (The $P_{1,{\rm M}}$ value given here for a scattering angle of $+10°$ should not be confused with the corrected P_1 value given in table 1.) The second check involved a measurement of the differential elastic scattering cross section. Before this measurement the alignment of the incident electron beam was checked. Next, the scattered electron count rate from the analyser channeltron was monitored for 30 s for a variety of scattering angles. Figure 1 shows the results together with measurements from Williams and Willis (1975) taken with significantly better angular resolution. Very good agreement in the position of the minimum at 71° is indicated. Both checks indicate that our electron scattering angles are determined with an accuracy of about 1°.

Table 1. Ar $4s'[\frac{1}{2}]$ Stokes parameters at 30 eV impact energy.

Scattering angle (deg)	P_1	P_4	ρ_{00}
10	−0.18±0.05	1.08±0.11	−0.016±0.022
15	−0.76±0.05	1.17±0.27	−0.009±0.014
20	−1.15±0.05	0.41±0.33	−0.032±0.029
25	−0.99±0.05	0.65±0.16	0.001±0.005
30	−0.22±0.05	1.09±0.19	−0.017±0.035
40	0.85±0.09	0.99±0.13	0.007±0.058
50	1.01±0.14	1.06±0.19	−0.028±0.095
60	0.87±0.11	1.17±0.18	−0.078±0.084
70	−0.27±0.11	1.05±0.24	−0.009±0.043

3. Theory

In the first excited states of the noble gases configuration mixing occurs and their wavefunctions are commonly written (e.g. Becker *et al* 1992) as a linear combination of pure Russell–Saunders states:

$$\begin{aligned} |ns'[\tfrac{1}{2}]_1^0\rangle &= c_0\,|{}^1\mathrm{P}_1\rangle + c_1\,|{}^3\mathrm{P}_1\rangle \\ |ns[\tfrac{3}{2}]_1^0\rangle &= -c_1\,|{}^1P_1\rangle + c_0\,|{}^3P_1\rangle \end{aligned} \tag{4}$$

where the mixing coefficients for argon are $c_0=0.893$ and $c_1=-0.450$. Correspondingly the scattering amplitudes for these states can be written as

$$f(M_1, m_1, m_0) = c_0 f_0(M_1, m_1, m_0) + c_1 f_1(M_1, m_1, m_0) \tag{5}$$

where $f_0(M_1, m_1, m_0)$ is a pure singlet amplitude and $f_1(M_1, m_1, m_0)$ is a pure triplet amplitude (Bartschat and Madison 1987). With the quantization axis chosen along the

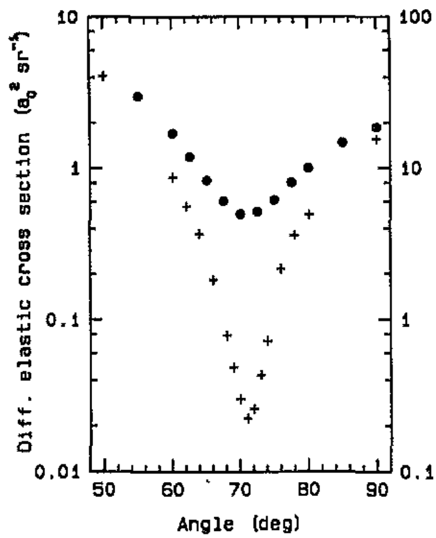


Figure 1. Circles, present data: scattered electron count rate as a function of scattering angle. Plusses: absolute differential elastic cross section for argon atoms from Williams and Willis (1975). Both measurements are at 30 eV incident electron energy. (See text for discussion.)

incident electron beam, ρ_{00} becomes:

$$\rho_{00} = \langle 1|\rho|1\rangle + \langle 1|\rho|-1\rangle \tag{6}$$

in terms of the reduced density matrix elements:

$$\langle M_1'|\rho|M_1\rangle = \frac{1}{2}\sum_{m_1 m_0} f(M_1', m_1, m_0) f^*(M_1, m_1, m_0). \tag{7}$$

Bartschat and Madison (1987) discuss the application of the non-relativistic approximation to the description of the scattering process. In this approximation the relativistic effects for the continuum electron are ignored and it is assumed that there are no spin-dependent forces in the scattering operator *T*. In this approximation the singlet and triplet amplitudes $f_s(M_1, m_1, m_0)$ can be expressed as linear combinations of their non-relativistic counterparts $f_s(M_1)$ (see Bartschat and Madison 1987, equation (13)). The direct and exchange cross sections can now be expressed as

$$\sigma_s = \frac{1}{2S+1}(|f_s(0)|^2 + 2|f_s(1)|^2) \tag{8}$$

and the differential cross sections become:

$$\frac{d\sigma}{d\Omega}[\tfrac{1}{2}] = c_0^2\sigma_0 + c_1^2\sigma_1 \tag{9}$$

$$\frac{d\sigma}{d\Omega}[\tfrac{3}{2}] = c_1^2\sigma_0 + c_0^2\sigma_1 \tag{10}$$

Similarly, the following expressions for ρ_{00} can be found:

$$\rho_{00}[\tfrac{1}{2}] = \frac{1}{2}\left[1 + \left(\frac{c_0}{c_1}\right)^2 \frac{\sigma_0}{\sigma_1}\right]^{-1} \tag{11}$$

$$\rho_{00}[\tfrac{3}{2}] = \frac{1}{2}\left[1 + \left(\frac{c_1}{c_0}\right)^2 \frac{\sigma_0}{\sigma_1}\right]^{-1}. \tag{12}$$

Thus, even in the absence of spin-dependent and/or relativistic forces, non-zero ρ_{00} values are expected in the case of the noble gases, where configuration interaction is relevant.

From these equations it is seen that the ratio of the exchange and direct cross sections can be obtained in two ways: (*a*) from a measurement of ρ_{00} as a function of scattering angle, and (*b*) from a measurement of the differential cross sections. Thus from equations (9 and 10) we find:

$$\frac{\sigma_1}{\sigma_0} = \frac{-c_1^2\dfrac{d\sigma}{d\Omega}[\tfrac{1}{2}] + c_0^2\dfrac{d\sigma}{d\Omega}[\tfrac{3}{2}]}{c_0^2\dfrac{d\sigma}{d\Omega}[\tfrac{1}{2}] - c_1^2\dfrac{d\sigma}{d\Omega}[\tfrac{3}{2}]} \tag{13}$$

and, also, from equations (11) and (12) we obtain

$$\frac{\sigma_1}{\sigma_0}=\left(\frac{c_0}{c_1}\right)^2\left[\frac{1}{2\rho_{00}[\frac{1}{2}]}-1\right]^{-1} \tag{14}$$

and

$$\frac{\sigma_1}{\sigma_0}=\left(\frac{c_1}{c_0}\right)^2\left[\frac{1}{2\rho_{00}[\frac{3}{2}]}-1\right]^{-1}. \tag{15}$$

Significantly, this allows a direct comparison to be made between coincidence measurements, yielding ρ_{00}, and non-coincidence measurements of the sublevel differential cross sections. Such a comparison is given in the next section.

4. Results and discussion

The data obtained for the excitation of Ar $4s'[\frac{1}{2}]^0_1$ at 30 and 20 eV impact energy are listed, for convenience, in tables 1 and 2 and displayed in figures 2 and 3. Also shown in the figures are three theoretical data sets due to da Paixao *et al* (1984), Bartschat and Madison (1987) and Zuo *et el* (1992). All three theories are first-order perturbative in nature and represent various distorted-wave approximations. da Paixao *et al* used the first-order many-body theory (FOMBT) of Csanak *et al* (1971). This is a form of the distorted wave approximation in which the distortion potential for both the incident and scattered electrons is the static-exchange field of the ground state. Spin-orbit effects in the target are included. Bartschat and Madison (1987) used a first-order distorted wave Born approximation (DWBA) and included relativistic effects in the description of the target states and in the wavefunction of the continuum electron. They found that they got best agreement with existing experimental data when the distorting potential for both the incoming and outgoing electron was the excited state Hartree potential plus a dynamic exchange potential. Zuo *et al* (1992) used a completely relativistic version of the distorted wave approximation (RDW) which had been introduced by Zuo *et al* (1991) in connection with e–Xe scattering. This automatically takes account of all effects involving electron spin. Following Bartschat and Madison (1987) they used the static potential of the final channel as the distortion potential for both the initial and

Table 2. Ar $4s'[\frac{1}{2}]$ Stokes parameters at 20 eV impact energy.

Scattering angle (deg)	P_1	P_4	ρ_{00}
15	0.21 ± 0.13	1.19 ± 0.44*	−0.06 ± 0.12
20	−0.64 ± 0.09	0.88 ± 0.23*	0.012 ± 0.023
25	−1.06 ± 0.15	−0.26 ± 0.68*	−0.06 ± 0.17
30	−1.02 ± 0.12	0.34 ± 0.36	−0.006 ± 0.030
35	−0.48 ± 0.17	1.09 ± 0.38	−0.011 ± 0.046
40	0.44 ± 0.09	1.14 ± 0.15	−0.048 ± 0.051
50	0.93 ± 0.16	1.03 ± 0.19	−0.015 ± 0.092
60	0.41 ± 0.13	0.96 ± 0.16	0.015 ± 0.058
70	0.28 ± 0.14	0.90 ± 0.24	0.032 ± 0.079
80	−0.36 ± 0.22	0.66 ± 0.63	0.06 ± 0.13

* Corrected for the $P_4[\frac{3}{2}]$ contribution using theoretical data, see discussion in section 2.

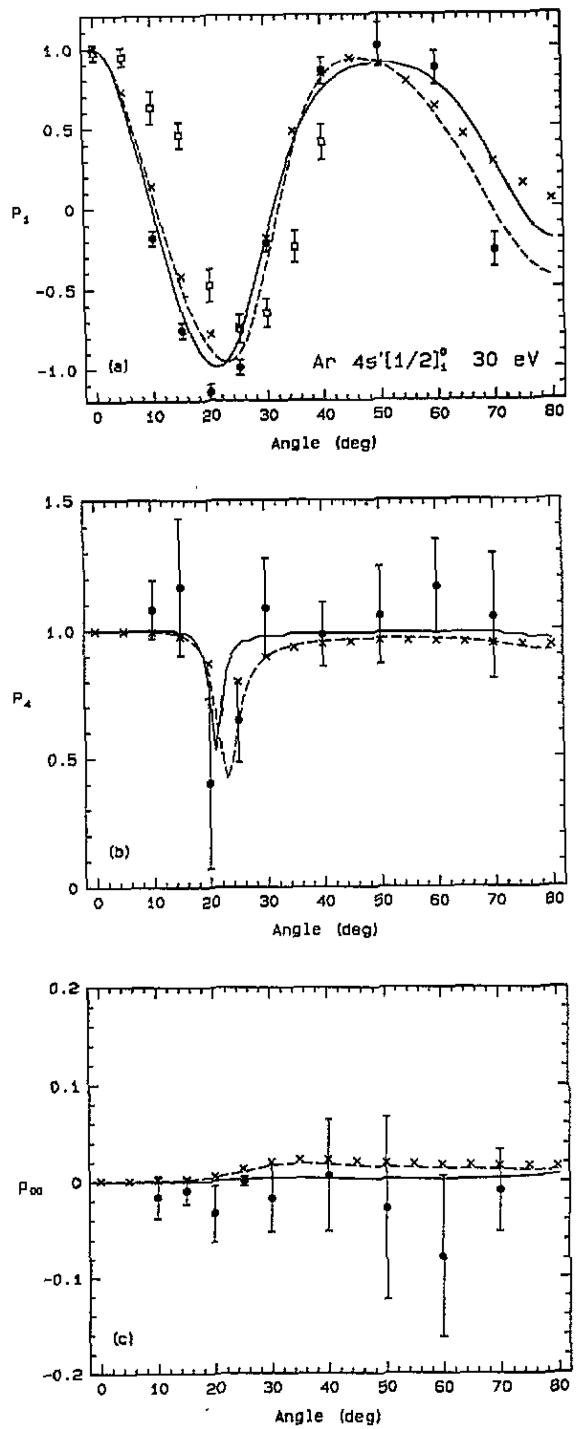
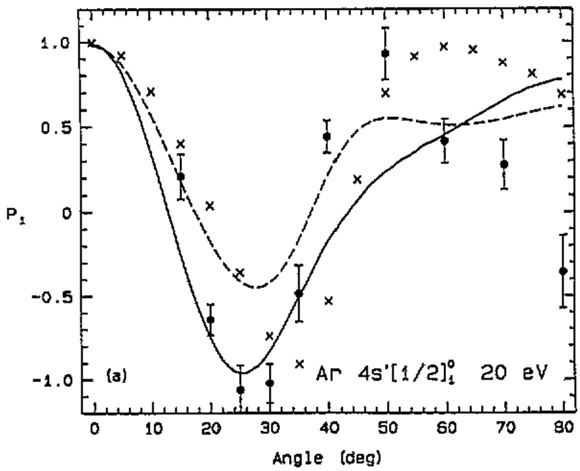
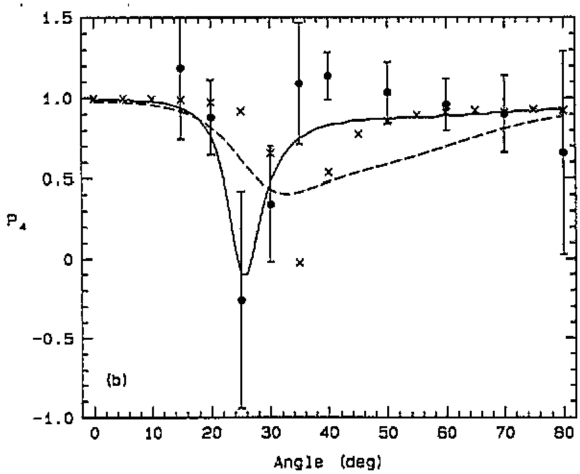


Figure 2. Variation of the P_1, P_4 and ρ_{00} parameters with electron scattering angle for excitation Ar $4s'[\frac{1}{2}]_1^0$ at 30 eV incident energy. Circles with error bars, present data; squares with error bars, experimental data from Zheng and Becker (1992); broken curve, RDW calculation of Zuo *et al* (1992); full curve, DWBA calculation of Bartschat and Madison (1987); crosses, FOMBT calculation of da Paixao *et al* (1984).





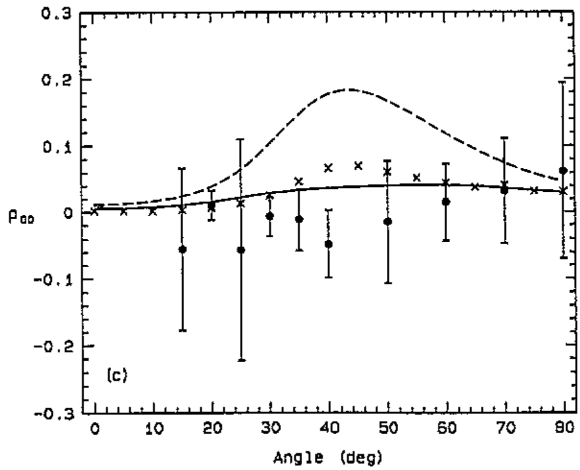


Figure 3. Variation of the P_1, P_4 and ρ_{00} parameters with electron scattering angle for excitation of Ar $4s'[\frac{1}{2}]_1^0$ at 20 eV incident energy. Symbols as in figure 2.

final channel distorted waves. This was found to give the best agreement with existing experimental data.

Looking at the 30 eV data in figure 2 we observe good agreement between our results and all the available theoretical calculations. The only discrepancy occurs in the P_l parameter at scattering angles greater than 60° where our P_l values are significantly lower than any of the theoretical calculations. A matter of some concern is the discrepancy between our data and the measurements of Zheng and Becker (1992). The minimum in their P_l measurements is -0.75 and occurs at around 25°, whereas the minimum in our data is -1 and occurs at around 20°. A possible reason for this discrepancy could be a systematic error in the calibration of the scattering angle in either experiment. As discussed earlier, we believe that our scattering angles are accurate to within 1°. We note that all the theories predict the rather narrow dip in the P_4 data, in the 20°–25° region. Our ρ_{00} values in figure 2(*c*) have error bars that are too large to reveal any small non-zero values, and do not allow us to comment on the small differences between the calculations which are evident at certain angles.

Comparison of the present P_l measurements at 20 eV, figure 3(*a*), with theory is of interest. The minimum in our P_l measurements at 30° seems to favour the DWBA calculation over the other two calculations, but at scattering angles between 50° and 80° none of the calculations matches our measurements. Also in case of the P_4 measurements, figure 3(*b*), the best agreement is with the DWBA calculation. The RDW and FOMBT calculations at 35° and 40° yield P_4 values that are noticeably lower than the experimental values. At 20 eV our ρ_{00} measurements, figure 3(*c*), have error bars that are too large to reveal any small non-zero values. There is good agreement with the DWBA and FOMBT calculations but the RDW calculation produces ρ_{00} values that seem to be too large.

In section 3 we obtained expressions (equations (4) and (5)) relating the ratio, σ_1/σ_0, of the exchange and direct cross sections to the ρ_{00} parameter. These expressions were derived under the assumption that relativistic effects involving the continuum electron could be ignored and that other spin related effects in the scattering process apart from exchange could also be ignored. Clearly in the present instance where all of our ρ_{00} measurements are zero within the experimental uncertainty it is not meaningful to evaluate σ_1/σ_0. However it is of interest to use the data of Corr *et al* (1992) and evaluate σ_1/σ_0 for the $4s[\frac{3}{2}]_1^0$ excitation process. The results of this are displayed in figures 4 and 5 for 30 and 20 eV excitation respectively. Also shown in the figures are the data of Chutjian and Cartwright (1981) making use of equation (13). Clearly good agreement, with regard to the trends in the data, is obtained. The two distinct peaks in the 20 eV data, figure 5, at 40° and 80° are clearly evident in both experimental data sets. The rather good agreement between the two data sets suggests that the assumptions, under which equations (11)–(15) were derived, are in fact justified. Thus relativistic effects involving the continuum electron and spin-dependent forces in the scattering operator can possibly be ignored for argon. It is an interesting challenge to theoreticians to suggest test cases where such effects would not be negligible and which might allow the various interactions to be unabigiously identified and studied.

The non-zero σ_1/σ_0 ratio and $\rho_{00}[\frac{3}{2}]$ values indicate that the $\rho_{00}[\frac{1}{2}]$ values should be non-zero as well. As an example, suppose that $\sigma_1/\sigma_0=0.20$ at a certain energy and scattering angle. Assuming the non-relativistic approximation is valid, we obtain from equations (11) and (12) the corresponding values $\rho_{00}[\frac{3}{2}]=0.22$ and $\rho_{00}[\frac{1}{2}]=0.024$. The uncertainties in our ρ_{00} measurements are such that a value of 0.22 would be readily verified experimentally, whereas a value of 0.024 would be obscured by the experimental

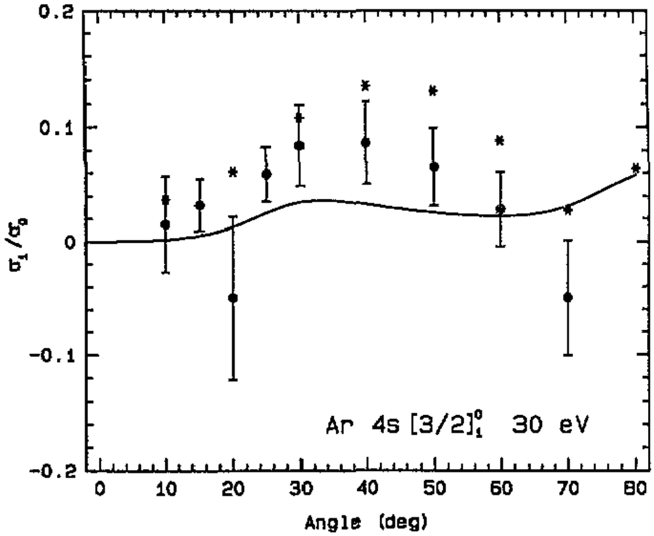


Figure 4. Ratio of exchange to direct cross sections as a function of electron scattering angle for excitation of Ar $4s[\frac{3}{2}]^o_1$ at 30 eV impact energy. Full circles, data from Corr *et al* (1992) using equation (15). Stars, data from Chutjian and Cartwright (1981) using equation (13). Full curve, DWBA data.

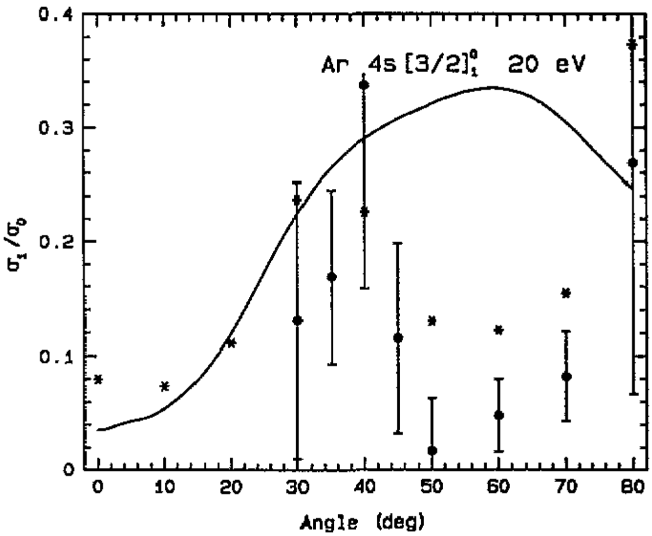


Figure 5. Ratio of exchange to direct cross sections as a function of electron scattering angle for excitation of Ar $4s[\frac{3}{2}]^o_1$ at 20 eV incident energy. Symbols as in figure 4.

error bars. This example illustrates that non-zero values for $\rho_{00}[\frac{1}{2}]$ are to be expected, but are so small that they are obscured by the experimental error bars.

Theoretical (DWBA) data are also shown in figures 4 and 5 for comparison. Although reasonable agreement with experiment is observed for the 30 eV data, this is not the case at 20 eV. This reflects the disagreement between experiment and theory for ρ_{00} data previously noted by Corr *et al* (1992). Given the fact that perturbative theories are not expected to apply at energies below a few times the threshold energy, the overall level of agreement between experiment and theory can be regarded as satisfactory.

5. Conclusions

Previous studies of the coherent excitation of the resonance states of argon have been extended to the 4s′ $[\frac{1}{2}]_1^0$ state for incident electron energies of 20 and 30 eV. Quite good agreement, between measured P_1, P_4 and ρ_{00} parameters and calculated values using various distorted wave theories, is obtained at 30 eV but at 20 eV significant discrepancies are observed as might be expected. Within the experimental error bars the ρ_{00} parameter was found to be zero at both energies and at all scattering angles studied. Previous data obtained for the 4s$[\frac{3}{2}]_1^0$ state has been analysed to give the ratio of the exchange to direct scattering cross sections for that state. Good agreement between this ratio and that obtained from non-coincidence differential cross section measurements is demonstrated.

Acknowledgments

The financial assistance of the Natural Sciences and Engineering Research Council of Canada is gratefully acknowledged as are helpful discussions with K Bartschat, K Becker, A Crowe, T H Gay and A D Stauffer. We are also grateful to D H Madison and A D Stauffer for supplying us with their tabulated coherence data. Finally, we thank Werner Grewe, Bernard Masse and the rest of the technical staff in the mechanical and electronic shops at Windsor for their expert and continued assistance.

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