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The total cross section for low-energy electron scattering from krypton

Stephen J Buckman and Birgit Lohmann†

Electron Physics Group, Atomic and Molecular Physics Laboratories, Research School of Physical Sciences, Australian National University, Canberra, ACT, Australia

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Abstract. Absolute total scattering cross sections have been measured for electron scattering from krypton in the energy range 0.175-20 eV. The measurements have been performed with a linear time of flight electron transmission spectrometer. Comparisons are made with the results of a number of other experiments and theoretical calculations and the agreement is generally good. However, at low energies, particularly in the region of the Ramsauer minimum, the present results and the recent measurements of Jost *et al* differ by more than 30%. A momentum transfer cross section is derived from the data by the use of the modified effective-range theory to enable direct comparison with swarm-derived cross sections.

1. Introduction

The increasing accuracy with which absolute low-energy total scattering cross sections have been measured in recent years has established such measurements as an effective first-order test of theoretical approaches to the electron-atom (molecule) scattering process. This paper is the second in a series on low-energy electron rare-gas scattering cross sections where particular emphasis is placed on scattering at incident energies below a few eV. In the heavier rare gases (Ar, Kr, Xe, Rn) the cross section in this energy range is dominated by the Ramsauer-Townsend (RT) minimum and the amount of reliable experimental data available for these atoms, in general, decreases with increasing atomic number.

There have been a number of experimental studies of electron scattering from krypton below 20 eV since the initial measurements of Ramsauer (1923) and Ramsauer and Kollath (1929). Total scattering cross sections (σ_t) have been measured by Gus'kov *et al* (1978), Dababneh *et al* (1980, 1982), Wagenaar and de Heer (1980, 1985) and Jost *et al* (1983) whilst total *elastic* cross sections have been derived from elastic differential cross sections by Srivastava *et al* (1981). Frost and Phelps (1964) obtained a momentum transfer cross section (σ_m) from measured electron drift velocities in the energy range 0.01-20 eV. A large number of calculations for electron-krypton elastic scattering have been carried out (see, for example, Walker 1971, McCarthy *et al* 1977, Yau *et al* 1980, Sin Fai Lam 1982, Kemper *et al* 1983, Berg 1983, Fon *et al* 1984, McEachran and Stauffer 1984, Haberland *et al* 1986), but only Yau *et al*, Sin Fai Lam,

† Permanent address: School of Mathematical and Physical Sciences, Murdoch University, Murdoch, WA, Australia.

Fon *et al* and McEachran and Stauffer have placed any particular emphasis on the energy region below 2 eV.

2. Experimental apparatus and procedures

The present measurements have been performed with a time of flight (TOF) spectrometer, which has been described in detail in a previous paper (Buckman and Lohmann 1986). In brief, an electron beam with an energy of, typically, 150 eV is pulsed and retarded in energy by a series of electrostatic lenses before entering a long (255 mm) attenuation cell. Those electrons which do not suffer a collision with the target gas within the cell and pass through the exit aperture are re-accelerated and focused into a channel electron multiplier detector. The absolute energy of the electrons within the scattering cell is determined from their TOF, and the total cross section determination at any particular energy involves measuring the transmitted electron flux both with and without krypton in the scattering cell and applying the Lambert-Beer relationship. The absolute gas pressure in the cell is measured by a spinning rotor viscosity gauge and the cell temperature is monitored by platinum resistance thermometers. The effective scattering length of the cell is taken to be the physical length.

The final measured cross sections represent the weighted mean of a number (at least 5 and as many as 16) of measurements at each energy which were performed over a wide range of experimental parameters such as electron optical settings and gas number density. The absolute uncertainties in the cross sections lie within 3-3.5% at all energies. A full description of the experimental uncertainties is given in Buckman and Lohmann (1986).

3. Results and discussion

The present total cross section is given in table 1 and a comparison with other measurements and theory is made in figure 1. At lower energies (<2 eV) the present results are in reasonably good agreement with the measurements of Ramsauer and Kollath (1929) and with those of Gus'kov *et al* (1978), particularly at energies below the RT minimum, which the present experiment locates at 0.74 eV. At 0.3 eV the present value is in excellent agreement with that of Jost *et al* (1983) but large differences are evident at higher energies, particularly, in the region of the minimum. While there is reasonable agreement in the position of the minimum (Jost *et al* (1983) locate it at 0.7 eV), the absolute magnitude of the cross section minimum in the present results is some 36% lower than that of Jost *et al* (1983). This discrepancy diminishes as the energy increases (e.g. 17% at 3 eV), until at energies above 7 eV the two data sets agree within combined error limits. This discrepancy will be discussed in some detail later.

At higher energies several other experimental cross sections are available for comparison. The measurements of Dababneh *et al* (1980) are in good agreement with the present results in the energy range of overlap (1.9-20 eV). Srivastava *et al* (1981) derived their total *elastic* cross section by extrapolating and integrating their differential elastic scattering cross sections for a number of energies between 3 and 100 eV to obtain cross sections with an estimated uncertainty of 30%. We have limited the comparison with their data to energies of 10 eV or less where the total elastic cross

Table 1. Total scattering cross sections for electrons in krypton.

Energy (eV)	σ_t (10^{-16} cm^2)	Error (1 SD %)
0.175	5.03	3.0
0.20	4.38	3.0
0.225	3.77	3.0
0.25	3.22	3.0
0.275	2.67	3.0
0.30	2.31	3.0
0.35	1.75	3.0
0.40	1.30	3.0
0.45	1.04	3.0
0.50	0.828	3.0
0.55	0.649	3.1
0.60	0.570	3.1
0.65	0.495	3.1
0.70	0.455	3.1
0.72	0.450	3.4
0.74	0.441	3.1
0.76	0.451	3.1
0.80	0.458	3.0
0.85	0.482	3.0
0.90	0.530	3.0
0.95	0.600	3.0
1.00	0.672	3.0
1.10	0.853	3.1
1.20	1.07	3.1
1.30	1.32	3.0
1.40	1.54	3.0
1.50	1.84	3.0
1.75	2.23	3.0
2.00	3.02	3.0
2.25	3.83	3.0
2.50	4.68	3.0
3.00	6.36	3.0
3.50	8.24	3.0
4.00	10.10	3.0
4.50	12.04	3.0
5.00	14.08	3.0
6.00	18.03	3.0
7.00	21.67	3.0
8.00	24.65	3.0
9.00	26.20	3.0
10.0	27.04	3.0
11.0	27.27	3.0
12.0	27.29	3.0
13.0	26.57	3.0
14.0	26.14	3.0
15.0	25.40	3.0
16.0	24.59	3.0
18.0	23.34	3.0
20.0	22.29	3.0

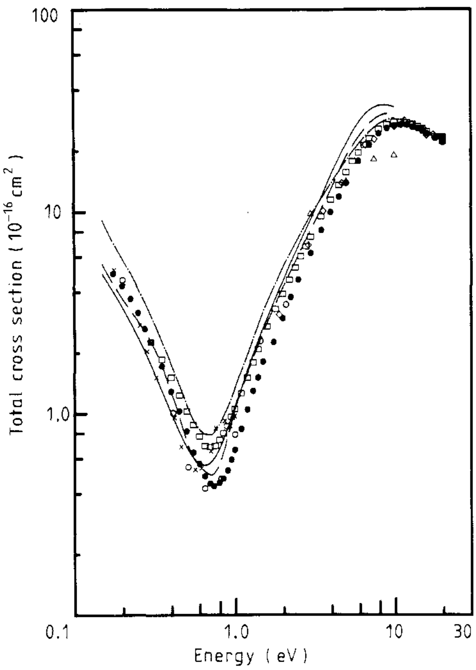

Figure 1. Total scattering cross sections for electrons in krypton: ●, present results; □, Jost *et al* (1983); ○, Gus'kov *et al* (1978); ▽, Dababneh *et al* (1980); ×, Ramsauer and Kollath (1929); △, Srivastava *et al* (1981); ■, Wagenaar and de Heer (1985); - - -, Sin Fai Lam (1982); — · —, Fon *et al* (1984); ——, McEachran and Stauffer (1984).

section is equivalent to the total cross section, the first excited-state threshold being 9.915 eV (Moore 1971). The energy dependence of their cross section between 3 and 10 eV is somewhat different from the present results. For example their 3 eV data point lies more than 50% above the present result whilst at 10 eV their value is 28% lower than the present result. This disagreement may well be a reflection of the difficulty involved in obtaining accurate integral scattering cross sections from differential measurements particularly when a large extrapolation to forward and backward angles is involved.

The final comparison with other experimental results is that with the recent data of Wagenaar and de Heer (1985). The energy range of overlap is unfortunately restricted to only one point at 20 eV. Nevertheless the agreement is excellent, being well within the combined uncertainties of the two data sets.

Comparison is also made in figure 1 with several theoretical elastic scattering calculations at energies below 10 eV. Sin Fai Lam (1982) used a semirelativistic model with polarisation included to calculate elastic scattering cross sections from 0.01 to 30 eV. At energies below the RT minimum this calculation is in excellent agreement with the present results. At energies above 0.7 eV the calculated cross section shows the same energy dependence as the present results but is larger in magnitude, being in excellent agreement with the data of Jost *et al* (1983) between 1 and 4 eV. Fon *et al* (1984) used the *R*-matrix approach to calculate elastic scattering cross sections from 0.1 to 120 eV. Relativistic effects are not included in this calculation but polarisation, exchange and absorption effects are included by coupling the ground state to a ^{1}P pseudostate. It can be seen from figure 1 that the calculation, whilst being everywhere higher than all the experimental results, reproduces the general features of the cross section very well. At the minimum the absolute value of the present cross section differs from the *R*-matrix prediction by 45%. At 0.175 eV this difference is 30% and at 10 eV the difference is 8%. Yau *et al* (1980) calculated scattering cross sections in the full polarised-orbital approximation with the inclusion of exchange through a local approximation. The results of this calculation are not shown in figure 1 but an extension of it by McEachran and Stauffer (1984) in which electron exchange is treated exactly and only the dipole part of the polarisation potential is retained, is given for comparison. As with the other two calculations the cross section of McEachran and Stauffer shows good general agreement with the present experimental cross section but there are regions where the differences in absolute magnitude are significant. The RT minimum in this calculation is at about 0.6 eV and the maximum in the cross section, at about 8.5 eV, is several eV below the maxima in the present results and the other calculations. The recent calculation of Haberland *et al* (1986) (not shown in figure 1) can be compared with the present results at 5 and 10 eV. This calculation describes the scattering process through a Kohn-Sham-type one-particle potential and gives total elastic cross sections which are 8% higher and 13% lower than the present results at 5 and 10 eV respectively.

The discrepancy which exists between the present results and those of Jost *et al* (1983), particularly in the region of the RT minimum, clearly warrants some detailed investigation. The excellent agreement which exists between the two data sets at the lowest energy common to both (0.3 eV) and at high energies (>7 eV), indicates that the difference cannot be due to a constant systematic error, such as a pressure gauge calibration, in either experiment. One possibility to which we have given extensive consideration is that the present results are too low due to inadequate discrimination against forward elastic scattering. In the region of the RT minimum the s-wave contribution to the scattering amplitude is negligible and the small but finite cross section is due to contributions from higher-order partial waves. The elastic differential cross sections in this region are therefore likely to be significantly enhanced at forward angles, and in a transmission experiment such as the present (or that of Jost *et al* (1983)) the failure to discriminate against these electrons results in an additional contribution to the transmitted electron flux when gas is present in the scattering cell and a corresponding decrease in the measured cross section. This effect has been postulated by Ferch *et al* (1985a) and ourselves (Buckman and Lohmann 1986) as the possible cause of similar differences found in argon where three recent measurements (Ferch *et al*, Jost *et al*, Buckman and Lohmann) are in excellent accord as to the position and magnitude of the RT minimum but disagree with several earlier measurements that have inferior discrimination against forward scattering.

As in the case of argon we have made an estimate of the effect of forward elastic scattering on the absolute value of our cross section at the RT minimum. If we consider the limiting aperture against forward scattering to be the exit aperture of the scattering cell, then the solid angle that this aperture subtends with respect to the centre of the cell is 1.94×10^{-4} sr. Using the calculated value for the elastic differential cross section at 0° and 0.8 eV (Fon *et al* 1984), and assuming the cross section to maintain this value within the whole forward solid angle, we find the resultant effect on the measured total cross section to be less than 0.1%. As a further experimental test we were able to significantly reduce the effective forward scattering solid angle by operating the accelerating lens subsequent to the scattering cell at the same potential as the cell (see Buckman and Lohmann (1986) for a description of the electron optics). This resulted in extending the field-free region beyond the scattering cell, and the last aperture in this lens, which was 1 mm in diameter, was now the limiting aperture against forward scattering. The solid angle that this aperture subtended at the centre of the scattering cell was 4.2×10^{-5} sr. Although the transmitted electron intensity was drastically reduced a series of measurements was performed in the region of the RT minimum. No significant differences were noted between the measured total scattering cross sections and those obtained from the initial measurements.

Another possible cause of a smaller measured cross section is multiple scattering, whereby an electron which has undergone a collision and would not normally leave the scattering cell is scattered back into the forward direction by a second collision. Such an effect is pressure dependent and would manifest itself as a decrease in the measured cross section with increasing pressure. No such effects have been observed in the present data at any energy for the range of pressures used in this study (5×10^{-4}–3×10^{-3} mbar).

The other alternative which must be considered is that the data of Jost *et al* (1983) are too high in the energy range 0.5–6 eV. As these data have only been published in a preliminary form we are not in a position to make any critical assessment of the experimental technique used by these authors, but we do make the following observations. Firstly, we note the comments of Wagenaar and de Heer (1985), who observed that the data of Jost *et al* show a larger increase with decreasing energy towards the low end of the energy range of comparison with their data (20 eV). They ascribe this to the extrapolation technique used by Jost *et al* to overcome the effects of forward scattering. This technique involves determining the dependence of the measured cross section on the size of the collision cell exit aperture, the final cross section being obtained by an extrapolation to zero width of the aperture. Wagenaar and de Heer point out that this technique can overestimate the 'scattering in' effect if the differential cross section has a tendency to level off in the forward direction. As a result the extrapolated cross section may be too large. Similar problems are noted by them in the case of xenon. Jost (1986) finds these arguments unconvincing and indicates that in the worst case the upward correction applied to their krypton cross section in the region of the RT minimum was 15% whilst at 2 eV it was 4%. Secondly, a cross section which is too large could be the result of an impurity such as N_2. We feel this is a most unlikely situation as the level of impurity required to explain the present discrepancy is in excess of 5%. High-purity gases were used for both the present measurements and those of Jost *et al*, and in the present measurements two different samples of krypton were used with no significant effect on the magnitude of the cross section at the RT minimum.

In addition to the comparison with experimental and theoretical total cross sections, we have investigated the efficacy of using modified effective-range theory (MERT) (O'Malley 1963) firstly, to extend the present results to zero energy and thus obtain the scattering length A for e^--krypton scattering, and secondly, to derive a momentum transfer cross section for comparison with the swarm-derived cross section of Frost and Phelps (1964). The four-parameter MERT expansions for the scattering phaseshifts have been given by O'Malley (1963), Haddad and O'Malley (1982) and Ferch *et al* (1985a):

$$\begin{aligned}
\tan\eta_0 &= -Ak[1+(4\alpha/3a_0)k^2\ln(ka_0)] - (\pi\alpha/3a_0)k^2 + Dk^3 + Fk^4 \\
\tan\eta_1 &= (\pi/15a_0)\alpha k^2 - A_1 k^3 \qquad (1) \\
\tan\eta_L &= \pi\alpha k^2/[(2L+3)(2L+1)(2L-1)a_0]
\end{aligned}$$

where α is the dipole polarisability for krypton (16.8 au, Miller and Bederson 1977), k is the wavenumber in au, a_0 is the Bohr radius and A, A_1, D and F are the four parameters determined from a fit to either the total or momentum transfer cross sections which are given by

$$\sigma_t = \frac{4\pi}{k^2} \sum_{L=0}^{\infty} (2L+1) \sin^2 \eta_L \tag{2}$$

$$\sigma_m = \frac{4\pi}{k^2} \sum_{L=0}^{\infty} (L+1) \sin^2(\eta_L - \eta_{L+1}). \tag{3}$$

This approach has been used by various groups to provide a theoretical bridge between σ_t and σ_m measurements for e^- rare-gas scattering (O'Malley and Crompton 1980, Haddad and O'Malley 1982, Ferch *et al* 1985a, Buckman and Lohmann 1986). However, the energy range over which MERT is applicable, either as a means of deriving the scattering length or as a means of a comparison between σ_t and σ_m, has not been established. It has also been demonstrated that in some cases, such as electron scattering from helium, the energy dependence of the cross section was not strong enough to enable a unique determination of both the s- and p-wave phaseshifts using MERT (Buckman and Lohmann 1986).

In the case of krypton we have attempted to determine the range of validity of MERT by analysing theoretical values (Fon *et al* 1984) of σ_t and σ_m at energies below 1 eV. The method employed was to fit the published R-matrix total cross section with the appropriate MERT expression for energies between 0.1 eV and E_{max}, where E_{max} varied between 0.5 and 1.0 eV, and then use the fitting parameters to calculate the corresponding momentum transfer cross section and compare it with the published R-matrix σ_m. The best agreement between the MERT-derived σ_m and the calculated σ_m was obtained with an E_{max} of 0.5 eV, although even with this maximum value of the fitting range the agreement above 0.3 eV was poor. Similar calculations were carried out in helium, neon and argon by analysing theoretical cross sections of Nesbet (1979) (helium), McEachran and Stauffer (1985) (neon) and Bell *et al* (1984) (argon). These indicated that MERT could be used to accurately convert from σ_t to σ_m or vice versa for energies up to about 1 eV for helium (providing the p-wave phaseshift is fixed) and neon and about 0.5 eV for argon. The smaller energy range in the heavier gases argon and krypton is thought to be a result of the increased anisotropy in the differential scattering cross sections at low energies for these atoms. As the total and momentum transfer cross sections are sensitive to different regions of the differential cross section it appears that, as a means of transferring accurately from one to the other, MERT is limited to those low-energy regions where the differential cross section is reasonably isotropic.

The results of the MERT fit to the present krypton total cross section with an $E_{max} = 0.5$ eV are summarised in table 2, where we also make a comparison with a similar fit to the data of Jost *et al* (1983) and to the momentum transfer cross section of Frost and Phelps (1964). Whilst we have reservations about the applicability of MERT for cross section comparisons at energies above 0.3 eV it should provide a reasonable means for extrapolating the low-energy cross section to zero energy. From table 2 we can see that there is reasonable agreement between the scattering length derived from the present data ($-3.19a_0$) and that from the swarm-derived cross section ($-3.32a_0$). The fit to the σ_t values of Jost *et al* reveals a much smaller scattering length ($-2.43a_0$) and highlights the differences in the energy dependence of the two measured

Table 2. MERT parameters obtained from fitting experimental cross sections with $E_{max} = 0.5$ eV.

Parameter	This Work	Jost *et al* (1983)	Frost and Phelps (1964)
A/a_0	−3.19	−2.43	−3.32
A_1/a_0^3	12.12	11.15	13.38
D/a_0^3	184.75	210.21	154.14
F/a_0^4	−300.8	−469.8	−72.92
Energy range used for fit (eV)	0.175–0.5	0.30–0.5	0.01–0.5

total cross sections at energies below the RT minimum. As a further comparison, the scattering lengths obtained from a MERT fit to the cross sections of Sin Fai Lam and of Fon *et al* ($E_{max} = 0.5$ eV) were −3.34 au and −3.79 au respectively. A MERT fit to the total cross section of McEachran and Stauffer resulted in a scattering length of −3.08 au in excellent agreement with their published value of −3.10 au.

In figure 2 we compare the MERT-derived σ_m from both the present data and from the data of Jost *et al* with the tabulated σ_m values of Frost and Phelps (from Kieffer 1973). The agreement between the present σ_m and that of Frost and Phelps is excellent at energies below 0.3 eV while that of Jost *et al* lies well below both the other cross

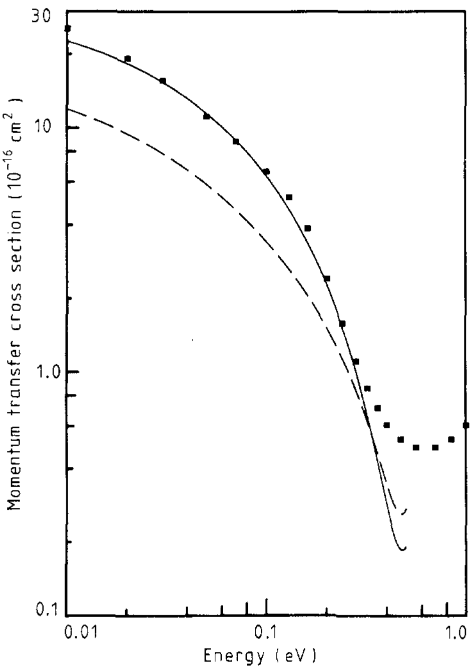

Figure 2. Momentum transfer cross section for krypton. ■, experimental values of Frost and Phelps (1964); MERT-derived cross sections: - - -, Jost *et al* (1983); ——, present results.

sections at energies less than 0.3 eV. At higher energies, particularly in the region of the RT minimum (at approximately 0.6 eV in σ_m), the swarm-derived cross section is larger than the cross section derived from the present results and from those of Jost *et al.* As this is the energy region where our reservations concerning MERT are strongest (it is the region of enhanced forward scattering), it is not appropriate to draw any firm conclusions from this discrepancy. However, since Frost and Phelps' work it has been shown that the swarm-derived cross section in the region of a RT minimum is constrained more closely by the lateral diffusion coefficient to mobility ratio, $D_\perp/\mu$, than by the drift velocity v_{dr} (Milloy *et al* 1977). The work of Milloy *et al*, based on measured data for both v_{dr} and $D_\perp/\mu$, gave a cross section for argon that was significantly smaller than that of Frost and Phelps in the region of the RT minimum. The cross section in argon measured by Frost and Phelps was obtained from drift velocity measurements only. It seems likely that a similar overestimation may have occurred in their krypton cross section at the RT minimum (Crompton 1986).

4. Conclusions

The total cross section presented here for electron–krypton scattering is in good general agreement with recent measurements and theoretical calculations. However, at energies below a few eV, and particularly in the region of the RT minimum at about 0.7 eV, the discrepancy between the present results and the other most recent experiment in this energy region, that of Jost *et al* (1983), is well outside the combined uncertainties in the two data sets. It is particularly baffling when one notes the excellent agreement obtained between cross sections measured with the present apparatus in the RT regions of argon and methane (Buckman and Lohmann 1986, Lohmann and Buckman 1986) and the measurements of Jost *et al* (1983) and Ferch *et al* (1985a, b). The reason for this discrepancy in krypton is not understood and clearly more experimental work in this area would be desirable. Unfortunately, similar differences exist between the three most recent theoretical calculations and as such they do not offer any assistance in resolving the problem.

Our efforts to make a comparison between total and momentum transfer cross sections obtained using modified effective-range theory indicate that the range of applicability of this approximation is somewhat less than in the lighter rare gases. We believe this to be a consequence of the increasing anisotropy of the low-energy differential scattering cross sections for the rare gases helium through to krypton. In the case of krypton, our analysis indicates that MERT is not valid as a means of comparing σ_t and σ_m for energies above about 0.3 eV.

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