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Elastic scattering of low energy electrons from sulphur dioxide

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Abstract. Absolute differential cross sections for vibrationally elastic electron scattering from sulphur dioxide (SO_2) have been measured using a crossed electron–molecular beam apparatus at incident energies from 1–30 eV. These data are compared with two previous experimental measurements and serve to resolve a discrepancy which existed in the absolute magnitude of these measurements. The differential cross section data have also been integrated to obtain total elastic and elastic momentum transfer cross sections.

1. Introduction

The study of low energy electron scattering from the sulphur dioxide (SO_2) molecule has attracted only limited experimental interest in recent years, despite its presence in the atmospheres of Jupiter, Io and Venus, in the interstellar medium and in our own atmosphere as one of the major pollutants. It has a relatively large permanent dipole moment (1.63 D) and as a result one may expect the low energy electron scattering cross sections to be large, as has been demonstrated in many other polar molecules (see Buckman *et al* 1994 for a brief review). Hence knowledge of these cross sections is also important in the modelling of discharge processes, such as diffuse discharge switches and lasers, where the molecule may occur, even as a trace impurity.

Previous experimental investigations include absolute total cross section measurements by Sokolov and Sokolova (1981) who base their experiment on an electron cyclotron resonance technique for the energy range of 1.5–10 eV, and by Zubek *et al* (1981) and Szmytkowski and Maciag (1986) who both employed a linear transmission experiment for the energy ranges 1.5–7 eV and 1.5–70 eV respectively. Zecca *et al* (1993) have recently applied the Ramsauer technique to the measurement of the total cross section at energies between 90 and 4000 eV. There have also been several measurements of total and partial ionization and attachment cross sections (for example Smith and Stevenson 1981, Cadez *et al* 1983, Orient and Srivastava 1983, 1984 and references therein) and of the production of metastable excited fragments following dissociation by electron impact (van der Burgt *et al* 1992). Hayashi (1987) has compiled an elastic momentum transfer cross section in the energy range of 0.3–1000 eV using available swarm and beam data and performing a consistency check on transport coefficients based on the Boltzmann equation in a Monte Carlo simulation method. Simon *et al* (1978) have measured excitation functions for various vibrational levels of the ground state of SO_2 in a study of the effects of the broad shape resonance centred at about

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3.4 eV. A more extensive set of measurement on these processes, including absolute differential cross sections for vibrational excitation of the ground state, were carried out by Andric *et al* (1983). The only absolute differential elastic cross sections available in the literature to this point, for the energy regime of the present study, are the measurements of Orient *et al* (1982) and Trajmar and Shyn (1989) at energies between 12–200 and 5–50 eV respectively. There are some substantial differences between these two measurements. While both measured cross sections show reasonably good agreement in shape at those energies where a comparision is possible, there appears to be a uniform difference of up to a factor of two in absolute magnitude. Trajmar and Shyn ascribe this difference to possible errors in the application of the relative flow technique in the work of Orient *et al*. Their own work does not use this technique to obtain the absolute magnitude but rather one based on measurements of relative scattering intensity for SO_2 and helium from a gas-flooded chamber, in conjunction with pressure measurements using a calibrated ion gauge.

There are no published theoretical calculations on SO_2 for the energy regime of the present work. However, the complex-Kohn variational technique, which has proved to be quite sucessful in the case of other polar molecules such as NH_3 and H_2S (see Buckman *et al* 1994), has recently been applied by Gil and Rescigno (1993) to SO_2. Thus the main incentive for the present work was to provide accurate, absolute experimental cross sections in an attempt to resolve the experimental differences at energies above 10 eV and to extend the impact energy range for which data is available to below 5 eV.

In the next section we discuss details of the experimental procedure employed in the current measurements, including a brief discussion of the normalization technique used for establishing the absolute scale. A discussion of our results is presented in section 3, with conclusions being drawn in section 4.

2. Experimental apparatus and procedures

The experimental measurements were carried out on a crossed electron–molecular beam apparatus. A detailed description of the apparatus and techniques can be found elsewhere (Brunger *et al* 1991, Gulley *et al* 1993) so only a brief discussion of the relevant features will be given here.

An incident beam of monoenergetic electrons produced from a conventional thermionic source was crossed with a beam of SO_2 effusing from a multicapillary array with an active diameter of 1 mm. Both the incident and scattered electrons were transported and energy resolved by a combination of electrostatic electron optics and a hemispherical electrostatic analyser. The scattered electron analyser rotated about the molecular beam enabling measurements at scattering angles between $-20°$ and $130°$. The true zero scattering angle was determined (to within $\pm 1°$) as that about which the intensity of the elastically scattered electrons was symmetric. A combination of Helmholtz coils and several layers of high permeability magnetic shielding reduced ambient magnetic fields in the region of the spectrometer to less than 1 mG. As we have described previously (Gulley *et al* 1993), the measurements were conducted with both gases present in the chamber at all times in an effort to minimize contact potential variations. This proved to be a great advantage as SO_2 is particularly aggressive, even at very low driving pressures, and several tungsten filaments were used during the duration of these measurements. The energy resolution for the present measurements

was about 40 meV. Whilst this was not sufficient to clearly resolve the first vibrational mode at an energy loss of 64 meV, the contribution of this state to the elastic intensity was negligible, even in the region of the 3.4 eV shape resonance where vibrational excitation is enhanced (Andric *et al* 1983). The incident energy was calibrated against the position of the structure in the N_2 ($v=0$–1) excitation function in the energy region of the N_2^- shape resonance (Ehrhardt and Willmann 1967) and against the position of the 2^2S He^- resonance at 19.367 eV (Brunt *et al* 1977).

The measured intensity of elastically scattered electrons from SO_2 was placed on an absolute scale by the use of the relative flow technique in conjunction with the helium elastic cross section. For energies less than 20 eV the theoretical cross section values of Nesbet (1979) were used for normalization, whilst at 20 and 30 eV we have used the measured values of Brunger *et al* (1992). The application of the relative flow technique, and some of the potential hazards involved, have also been discussed in detail before (Brunger *et al* 1991, Buckman *et al* 1993) and we will not repeat that discussion here. However the technique does require the accurate measurement of relative flow rates of both gases as a function of driving pressure.

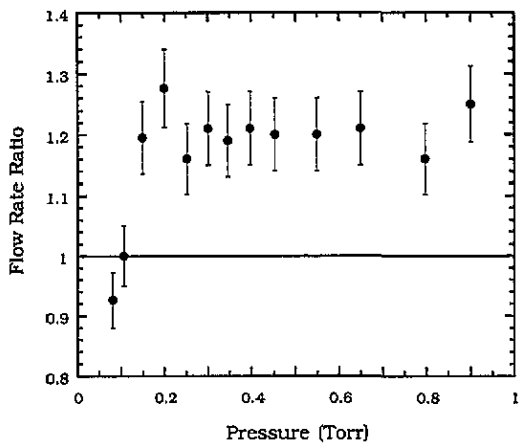

Figure 1. Ratio of the normalized flow rate ($F M^{1/2}$) for SO_2 to that of helium as a function of capillary array driving pressure.

These measurements are shown in figure 1 where they are presented as a ratio of the normalized flow rate (the flow rate (F) mulitplied by the root of the molecular mass (M)) for SO_2 to that of helium. Ideally this ratio should be unity but clearly, at most of the pressures studied here, it is not. Rather, the observed variation of this ratio from unity of 20% or more is typical of most such relative flow rate measurements and serves to highlight the necessity to actually measure these rates as accurately as possible. Typical driving pressures used in the present experiment were 0.2–0.4 Torr for SO_2 and 0.5–0.95 Torr for helium—the ratio of the pressures being determined by the requirement that the mean free paths for the two gases be the same at the entrance to the capillary array. These pressures were measured by an MKS Baratron capacitance manometer located upstream from the capillary array.

Ratios of scattered electron intensities for SO_2 and He, corrected for background contributions, were measured at each angle and the final cross sections represent a weighted average of many such measurements. The driving pressures and beam currents

were monitored continually during the measurement and the ratios normalized accordingly, although these parameters varied by typically less than 1% during a data collection cycle (30–80 min). Data accumulation was performed entirely under computer control.

3. Results and discussion

Absolute differential cross sections for elastic scattering by SO_2 at incident energies from 1.0–30 eV are given in table 1 and shown, at selected energies, in figures 2–6. The figures in brackets in the tables represent the absolute uncertainty (one standard deviation) expressed as a percentage. These arise from the combination of errors due to statistics, the measured relative flow rates, variations in pressure and beam current and the error associated with the helium cross section which is used for normalization.

At 1.0 eV (figure 2) the differential cross section exhibits the familiar strong forward scattering, the cross section rising to 20.7 $Å^2$ at an angle of 15°. In the backward direction ($\theta > 70°$) the DCS is rather flat with a relatively constant value of about 1 $Å^2$. There is no other experimental data or theory with which to compare at this energy.

Figure 3 shows the elastic DCS at an incident energy of 3.4 eV, corresponding to the mean energy of a broad shape resonance which was first detected by Sanche and Schulz (1973). Subsequent measurements by Simon *et al* (1978) and Andric *et al* (1983) have shown that this temporary SO_2^- state decays preferentially via the ν_1 $(n, 0, 0)$ vibrational mode. The detailed analysis of Andric *et al* further demonstrated that the symmetry of this state was most likely $^2B_2$ with the additional electron in a $6b_2$ orbital. Simon *et al* (1978) also measured excitation functions for elastic scattering and found evidence for decay of the resonance via the elastic channel. The present DCS for elastic scattering evolves from a rather flat backward angle behaviour at 1.0 eV to a deep minimum at 90° in the 3.4 eV DCS. Whilst the minimum persists at higher energies it once again flattens and moves to smaller angles. Such behaviour, masked as it is by the dominant dipole interaction, is consistent with the observed p-wave behaviour of the scattered electron in the vibrational excitation measurements of Andric *et al*. A similar, but much stronger, manifestation of shape resonance effects in elastic scattering has been observed in this laboratory in both NH_3 and H_2S (Alle *et al* 1992, Gulley *et al* 1993).

At an incident energy of 5 eV (figure 4) the elastic differential cross section has a noticeably different shape in the forward direction with a weak shoulder being evident between 20°–50°. At this energy we can also compare with the DCS measurements of Trajmar and Shyn (1989) and with the Kohn variational calculation of Gil and Rescigno (1993). This fixed nuclei approximation calculation was performed with a multiconfiguration, correlated target wavefunction. There is reasonable agreement between experiment and theory for both the shape and absolute magnitude of the DCS. The agreement with the experiment of Trajmar and Shyn is excellent at all but the most forward of scattering angles. At a scattering angle of 10° the present cross section is about 50% larger than that of Trajmar and Shyn.

Figure 5 illustrates the elastic DCS at an energy of 12 eV, the lowest energy at which comparison can also be made with the experiment of Orient *et al* (1982). The present cross section is once again in excellent agreement with that of Trajmar and Shyn but larger than that of Orient *et al* by between 30 and 100%. At this energy the results of Trajmar and Shyn are slightly higher (~20%) than the present ones at forward scattering angles. The Kohn calculation is in excellent agreement with the present cross section

Table 1. Absolute differential cross sections for elastic electron scattering from SO_2 in units of 10^{-16} cm^2 sr^{-1}. Figures in brackets represent the estimated uncertainty (%) at the 1 standard deviation level. Total elastic and elastic momentum transfer cross sections are also given in units of 10^{-16} cm^2. The uncertainty in these figures is estimated at ±20% for 5 eV and above and ±30% at energies below 5 eV. The figures in brackets represent the percentage of the integral cross section that arises from the extrapolated region.

	Energy (eV)								
Angle	1.0	2.0	3.4	5.0	10	12	15	20	30
10	—	—	—	—	—	18.98 (7.0)	19.05 (7.3)	20.19 (10.0)	23.82 (7.9)
12	—	—	—	12.67 (8.8)	—	—	—	—	—
15	20.70 (12.6)	16.92 (16.3)	13.45 (12.4)	9.60 (7.8)	13.27 (8.3)	—	14.74 (8.0)	—	14.82 (7.4)
20	12.89 (8.2)	10.16 (11.5)	8.31 (7.9)	7.00 (7.3)	9.71 (7.5)	10.69 (7.4)	10.94 (7.8)	9.80 (7.2)	9.65 (7.4)
25	—	—	6.10 (13.4)	5.42 (7.5)	—	—	8.57 (7.7)	—	5.97 (7.2)
30	6.63 (7.9)	5.33 (10.0)	4.55 (8.8)	4.55(7.5)	5.92 (7.3)	6.49 (8.0)	6.64 (7.5)	5.22 (7.2)	3.76 (7.4)
35	—	—	3.95 (7.7)	3.90 (7.3)	—	—	4.95 (7.5)	—	2.31 (7.5)
40	3.60 (7.4)	2.97 (8.1)	3.06 (7.99)	3.40 (7.4)	3.71 (7.9)	3.77 (7.3)	3.56 (7.5)	2.57 (7.3)	1.46 (7.2)
45	—	—	2.52 (8.4)	2.95 (7.2)	—	—	2.51 (7.4)	—	0.880 (7.5)
50	2.20 (7.3)	1.98 (8.0)	2.06 (7.9)	2.56 (7.3)	2.21 (7.5)	1.93 (7.5)	1.65 (7.4)	1.09 (7.2)	0.587 (7.4)
55	—	—	1.77 (9.5)	2.19 (7.3)	—	—	1.08 (7.9)	—	0.429 (7.2)
60	1.41 (8.5)	1.33 (8.1)	1.44 (9.5)	1.84 (7.4)	1.23 (7.7)	0.933 (7.3)	0.684 (7.5)	0.452 (7.5)	0.355 (7.3)
65	—	—	—	—	—	—	0.499 (7.5)	0.376 (7.3)	—
70	1.13 (8.2)	1.07 (8.0)	1.06 (8.8)	1.35 (7.6)	0.773 (7.3)	0.573 (7.5)	0.426 (7.5)	0.354 (7.7)	0.327 (7.4)
75	—	—	—	—	—	—	0.432 (7.5)	—	—
80	0.995 (8.4)	0.940 (7.4)	0.830 (9.4)	0.995 (7.2)	0.617 (7.3)	0.523 (7.2)	0.482 (7.7)	0.401 (7.3)	0.324 (7.7)
85	—	—	0.788 (12.7)	0.891 (7.5)	—	—	—	—	—
90	0.989 (7.5)	0.849 (9.7)	0.714 (7.6)	0.857 (7.3)	0.631 (7.4)	0.591 (7.3)	0.563 (7.4)	0.426 (7.3)	0.328 (8.0)
95	—	—	0.703 (7.5)	0.848 (7.4)	—	—	—	—	—
100	0.941 (8.8)	0.901 (7.5)	0.746 (7.6)	0.903 (7.3)	0.733 (7.3)	0.711 (7.4)	0.637 (7.4)	0.460 (7.7)	0.330 (7.6)
110	0.977 (7.8)	0.959 (8.3)	0.868 (8.1)	1.03 (7.2)	0.879 (7.3)	0.826 (7.5)	0.694 (7.3)	0.491 (7.3)	0.369 (8.0)
120	0.983 (8.0)	1.06 (7.5)	1.07 (8.3)	1.22 (7.2)	1.03 (7.2)	0.973 (7.3)	0.773 (7.3)	0.549 (7.6)	0.423 (7.3)
130	1.02 (7.9)	1.29 (7.5)	1.42 (8.6)	1.48 (7.3)	1.17 (7.4)	1.11 (7.3)	0.865 (7.5)	0.649 (7.6)	0.514 (8.9)
Q_T	37.9 (42%)	36.3 (48%)	32.1 (40%)	29.4 (34%)	28.1 (36%)	26.4 (27%)	24.2 (24%)	19.9 (26%)	17.0 (28%)
Q_M	15.1 (24%)	16.7 (31%)	18.5 (39%)	19.7 (45%)	14.5 (40%)	13.7 (41%)	11.4 (39%)	8.6 (40%)	6.8 (43%)

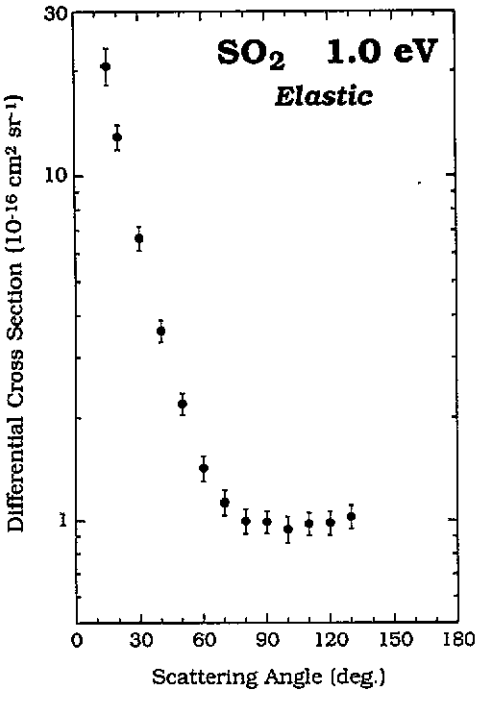

Figure 2. Differential cross section for elastic electron scattering from SO_2 at 1.0 eV in units of 10^{-16} cm^2 sr^{-1}.

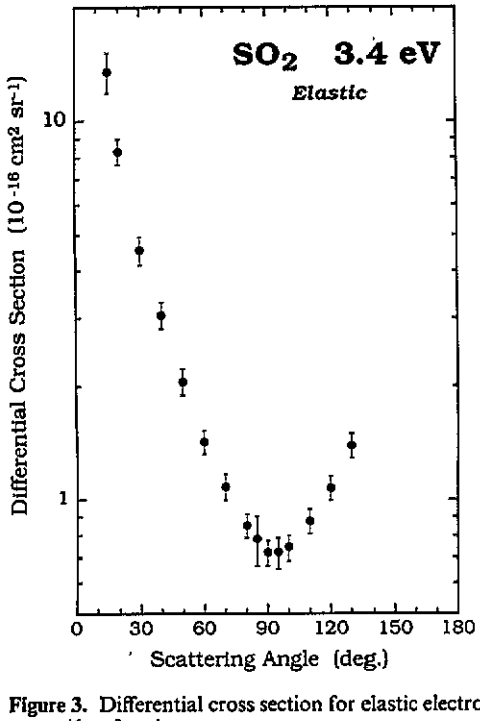

Figure 3. Differential cross section for elastic electron scattering from SO_2 at 3.4 eV in units of 10^{-16} cm^2 sr^{-1}.

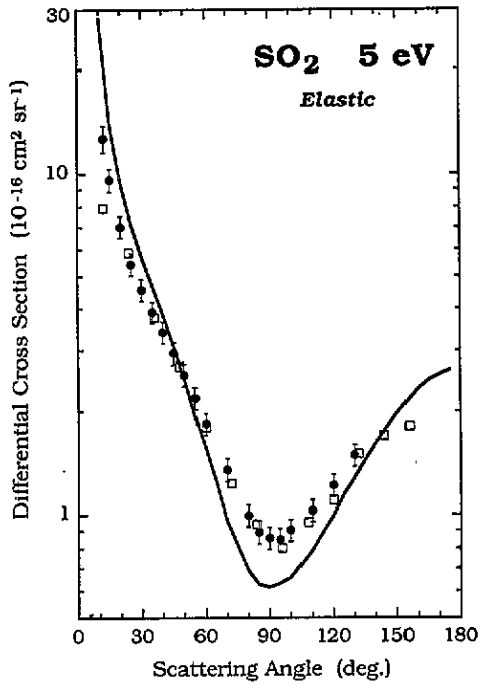

Figure 4. Differential cross section for elastic electron scattering from SO_2 at 5.0 eV in units of 10^{-16} cm^2 sr^{-1}. Present data (●), Trajmar and Shyn (□), Gil and Rescigno (——).

for angles less than about 50°, and whilst it predicts the overall shape of the cross section rather well, it lies above the experiment for angles out to 130°.

A final comparison of the DCS is made at an incident energy of 20 eV (figure 6). Once again there is good agreement between the present experiment and that of Trajmar and Shyn, although at this energy there are small differences (10–30%) at backward angles ($\theta > 110°$). The data of Orient *et al* are also once again lower than the other experiments by between 20 and 150%. The Kohn calculation is in reasonable agreement with the present data and those of Trajmar and Shyn at forward angles but is once again higher at scattering angles larger than about 60°.

In the interests of brevity we do not show the DCS data at the other four measured energies. In general the trends which emerge from the data illustrated above are also evident at the other energies. The agreement with the Kohn calculation is reasonable at all energies although it consistently overestimates the backward scattering cross section as the energy is increased. Unlike the recent application of this approach to NH_3 and H_2S (see Rescigno *et al* 1992) the effects of open inelastic channels are not accounted for in the present work on SO_2 and, as the contribution from these open channels appears to be significant at energies above about 10 eV (see below), it may account for the theory overestimating the cross section at large angles, a trend that was not observed in either NH_3 or H_2S. The agreement with the experiment of Trajmar and Shyn is, in general, excellent at all energies with the exception of 30 eV, where the present cross section appears to be higher by between 10–50%. This difference cannot be due to the different helium cross sections used for the normalization. The present cross sections are also uniformly higher than those of Orient *et al* at all energies where a comparison is possible.

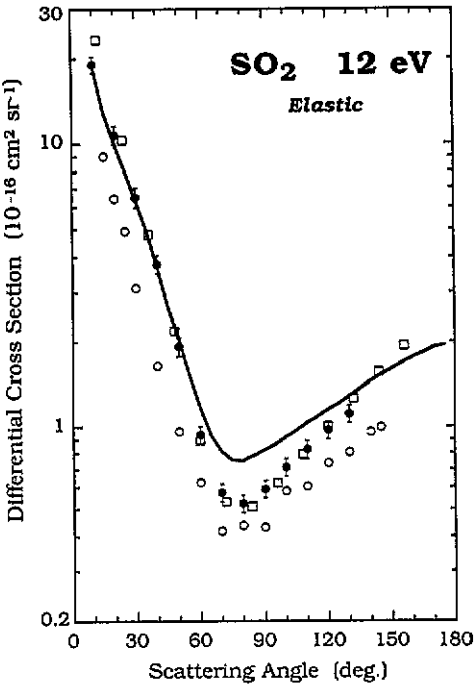

Figure 5. Differential cross section for elastic electron scattering from SO_2 at 12 eV in units of 10^{-16} cm^2 sr^{-1}. Present data (●), Orient *et al* (○), Trajmar and Shyn (□), Gil and Rescigno (——).

The present DCS have been extrapolated to forward and backward angles and integrated to yield both total elastic and elastic momentum transfer cross sections. At energies above 5 eV, the extrapolation has been carried out using the theoretical cross section as a guide to the shape of the DCS in those regions where we do not measure it. Below 5 eV this extrapolation has been done by eye. The resultant cross sections are also shown in table 1 together with a figure representing the percentage of the integral cross section which results from the extrapolated region. Given the magnitude of the contributions from these regions, particularly in the case of the momentum transfer cross section, and by varying the contributions in these regions within reasonable limits, we place a conservative uncertainty on the derived integral cross sections of ±20% for energies of 5 eV and above, and 30% for energies below 5 eV. Whilst this level of uncertainty is rather unsatisfactory it does highlight the difficulties involved in obtaining reliable integral cross sections for highly polar molecules, particularly from differental measurements. These cross sections are shown in figure 7(*a*) and (*b*) where they are compared with other available experimental cross sections and theory.

The total elastic cross section (figure 7(*a*)) can be compared with the derived values of Trajmar and Shyn and Orient *et al* as well as the measured grand total cross section of Szmytkowski and Maciag (1986) and the theoretical total elastic cross section of Gil and Rescigno. As expected, the present results lie below those of Szmytkowski and Maciag at all energies and, if one considers the effects of other possible inelastic scattering processes, they appear to be consistent. For example, at 20 eV, if we add the measured total ionization cross section of 2.0 Å^2 (Orient and Srivastava 1984) and the measured integral inelastic (excluding ground state vibrational excitation which should

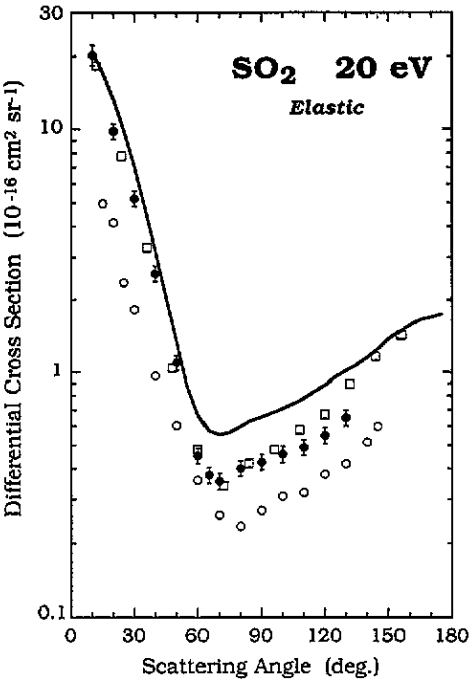

Figure 6. Differential cross section for elastic electron scattering from SO_2 at 20 eV in units of 10^{-16} cm^2 sr^{-1}. Present data (●), Orient *et al* (○), Trajmar and Shyn (□), Gil and Rescigno (——).

be negligible) cross section of 1.4 Å^2 (Vuskovic and Trajmar 1982) to the present total elastic cross section of 19.9 Å^2, the result is in excellent agreement with that of Szmytkowski and Maciag (23.0 Å^2). A similar exercise can be carried out at 3.4 eV where the only other significant scattering process is vibrational excitation. The measured total vibrational excitation cross section of Andric *et al* (1.7 Å^2), together with the present figure for elastic scattering (32.1 Å^2) is also in reasonable agreement with the measured total cross section of Szmytkowski and Maciag. It is interesting to note that there is no apparent structure in the grand total cross section, even at the energy of the 3.4 eV resonance, indicating the extent to which the scattering is dominated by the dipole interaction.

The elastic momentum transfer cross section is shown in figure 7(*b*) where it is compared with the derived cross section of Hayashi (1987), the integrated cross sections of both Orient *et al* and Trajmar and Shyn, and the theoretical cross section of Gil and Rescigno. Once again there is good agreement between the present experiment and that of Trajmar and Shyn. Both of these cross sections lie below that of Hayashi except at energies below 5 eV. The present cross section does appear to decrease for energies below 5 eV in a similar fashion to that of Hayashi and apparently the Kohn calculation, although this observation must be tempered by the considerations discussed above concerning the uncertainty on the derived integral cross sections. As might be expected from the comparison of the DCSs, the momentum transfer cross section of Gil and Rescigno is larger than the experimental values at all energies, with the exception of 5 eV.

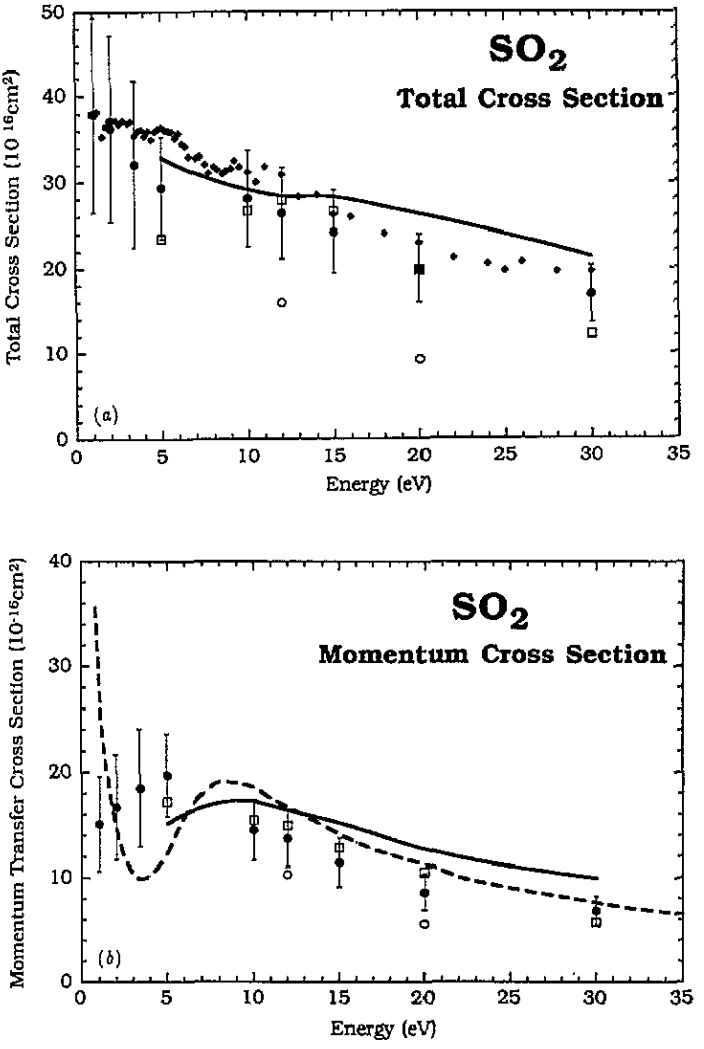

Figure 7. (*a*) Total elastic scattering cross section for SO_2 in units of 10^{-16} cm^2. Present data (●), Orient *et al* (○), Trajmar and Shyn (□), Szmytowski and Maciag (◆), grand total cross section, Gil and Rescigno (——). (*b*) Elastic momentum transfer cross section for SO_2 in units of 10^{-16} cm^2. Present data (●), Orient *et al* (○), Trajmar and Shyn (□), Hayashi (– – –), Gil and Rescigno (——).

4. Conclusions

The present measurements provide an extensive set of low to intermediate energy, absolute elastic differential cross sections which can be compared with a variety of experimental measurements and theoretical calculations at both the differential and total cross section level. In particular they provide the first such absolute measurements at energies below 5 eV. Comparison with the two previous measurements of differential scattering cross sections by Orient *et al* (1982) and Trajmar and Shyn (1989) confirms the discrepancy in absolute magnitude that was highlighted by the latter and adds weight to their hypothesis that the data of Orient *et al* suffer from (at least) an error in absolute normalization.

The comparison with the recent Kohn variational calculation (Gil and Rescigno 1993) is favourable at most energies although the theoretical calculation is higher than experiment at backward angles for most energies. This may be due in part to the relatively large inelastic cross sections in SO_2, which result in a loss of flux from the elastic channel, and which are not being accounted for in the calculation.

Acknowledgments

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