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On the excitation of the 2^3S, 3^3S and 4^3S states in helium by electron impact

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Abstract. A new plane wave approximation for calculating exchange scattering amplitudes is described and applied to the calculation of some singlet–triplet cross sections in helium. Comparison is made with experimental measurements of both total and differential cross sections. It is shown that a considerable improvement over previous techniques is obtained but that there remain significant differences between the theory presented and experimental measurement.

1. Introduction

The development of a procedure whereby exchange scattering amplitudes may be calculated simply and reasonably accurately has proved a persistent problem which appears to be considerably more difficult than the analogous problem of calculating direct scattering amplitudes. The earliest approximation due to Oppenheimer (1928) is very poor and while a number of alternative plane wave approximations have been advanced (for a recent review see Rudge 1973) none of them has proved satisfactory.

In a recent paper (Mathur and Rudge 1973) we examined a different method of calculating exchange scattering amplitudes in which only one partial wave occurred. This partial wave is one which is estimated quite wrongly in the Oppenheimer approximation and we showed that our procedure for its evaluation represented a great improvement as judged by low energy total exchange cross sections. In this paper we extend this treatment by adding the higher partial wave contributions as given by the Oppenheimer method which we anticipate to be less in error than the dominant contribution to this approximation for which we retain the procedure described previously. We compare the results thus obtained with experimental total and differential cross sections.

2. Theory

It is convenient to first develop the Oppenheimer amplitude in partial wave form. We adopt the same notation as that of Mathur and Rudge (1973) and set

$$I = -(2\pi)^{-1}\langle \Psi_q(-\boldsymbol{k}_q|\boldsymbol{r}_3, \boldsymbol{r}_1; \boldsymbol{r}_2)(\mathscr{H}-E)\Psi_p(\boldsymbol{k}_p|\boldsymbol{r}_1, \boldsymbol{r}_2; \boldsymbol{r}_3)\rangle \tag{1}$$

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where

$$\Psi_p(\boldsymbol{k}_p|\boldsymbol{r}_1, \boldsymbol{r}_2; \boldsymbol{r}_3) = \psi_p(LS|\boldsymbol{r}_1, \boldsymbol{r}_2) \exp i\boldsymbol{k}_p \cdot \boldsymbol{r}_3 \tag{2}$$

$$\Psi_q(-\boldsymbol{k}_q|\boldsymbol{r}_3, \boldsymbol{r}_1; \boldsymbol{r}_2) = \psi_q^*(L'S'|\boldsymbol{r}_3, \boldsymbol{r}_1) \exp -i\boldsymbol{k}_q \cdot \boldsymbol{r}_2. \tag{3}$$

The atomic wavefunctions can, as previously, be written in the form

$$\psi_i(LS|\boldsymbol{r}_1, \boldsymbol{r}_2) = (r_1 r_2)^{-1} \sum_{l_1 l_2} y(l_1 l_2 LM|\hat{\boldsymbol{r}}_1, \hat{\boldsymbol{r}}_2) X_i(l_1 l_2|r_1, r_2). \tag{4}$$

We define

$$\begin{aligned} \gamma_\lambda(r_1, r_2) &= \frac{r_1^\lambda}{r_2^{\lambda+1}} \qquad r_1 < r_2 \\ &= \frac{r_2^\lambda}{r_1^{\lambda+1}} \qquad r_1 > r_2 \end{aligned} \tag{5}$$

$$T_1 = \left\langle X_q(l_1' l_2'|r_3, r_1) j_{l'}(k_q r_2) \left(\gamma_\lambda(r_1, r_3) - \frac{1}{r_3}\right) X_p(l_1 l_2|r_1, r_2) j_l(k_p r_3) \right\rangle \tag{6}$$

$$T_2 = \left\langle X_q(l_1' l_2'|r_3, r_1) j_{l'}(k_q r_2) \left(\gamma_\lambda(r_2, r_3) - \frac{1}{r_3}\right) X_p(l_1 l_2|r_1, r_2) j_l(k_p r_3) \right\rangle \tag{7}$$

$$A_1 = (-1)^l W(L'lLl'; l_1 J) f_\lambda(l_1' l_2' l l_1 L') \delta_{l_2 l'} \tag{8}$$

$$A_2 = (-1)^{l_1'} W(L' l_1' L l_2; l_1 J) f_\lambda(l l' l_1' l_2 J) \delta_{l_2' l_1} \tag{9}$$

where W is a Racah coefficient and the coefficient f_λ is defined by Percival and Seaton (1957).

On expanding the plane waves in spherical harmonics and assuming that the atomic wavefunctions are exact we find that the integral of equation (1) reduces to

$$\begin{aligned} T = -\frac{8\pi}{k_q k_q}(2L'+1)^{1/2} &\sum_{ll'JM_3} i^l(-i)^{l'}(2J+1)^{1/2} C^{L'JL}_{M'M_J M} \\ &\times y(ll'JM_J|\hat{\boldsymbol{k}}_p, \hat{\boldsymbol{k}}_q) \sum_{l_1 l_2 l_1' l_2' \lambda} (A_1 T_1 + A_2 T_2) \end{aligned} \tag{10}$$

where C denotes a Clebsch–Gordon coefficient. If $\lambda = 0$ then it follows that $l = l_1'$, $l_2' = l_1$ and $l_2 = l'$. This term is spuriously large for transitions between s-states when evaluated by the Oppenheimer method. We interpret this as being due to the inadequacy of plane waves in the region where the free electron penetrates the atom. Our procedure for s–s transitions is therefore to take a regional trial function which corresponds to plane waves outside the atom for the partial waves which correspond to $\lambda = 0$ and to use plane waves everywhere for the other partial waves. We then obtain the result that

$$\begin{aligned} g_{pq}(\boldsymbol{k}_p, \boldsymbol{k}_q) = -\frac{8\pi(2L'+1)^{1/2}}{k_p k_q} &\sum_{ll'JM_J} i^l(-i)^{l'}(2J+1)^{1/2} C^{L'JL}_{M'M_J M} \\ &\times y(ll'JM_3|\hat{\boldsymbol{k}}_p, \hat{\boldsymbol{k}}_q) T(l, l', J) \end{aligned} \tag{11}$$

where

$$T(l, l', J) = \sum_{\substack{l_1 l_2 \\ l_1' l_2' \\ \lambda}} A_1[(1-\delta_{\lambda 0})T_1 + \delta_{\lambda 0} T_3] + (1-\delta_{\lambda 0}) A_2 T_2 \tag{12}$$

and

$$T_3 = -\frac{1}{2}\int_0^\infty \frac{\mathrm{d}}{\mathrm{d}r}(j_l(k_p r) j_{l'}(k_q r)) \int_0^r X_p(l_1 l'|r_1, r) X_q(l l_1|r, r_1)\,\mathrm{d}r_1$$
$$-\frac{1}{4}\int_0^\infty j_l(k_p r) j_{l'}(k_q r) X_p(l_1 l'|r, r) X_q(l l_1|r, r)\,\mathrm{d}r. \tag{13}$$

On averaging over the initial magnetic sub-states and summing over the final magnetic sub-states we find that the differential cross section is

$$\frac{\mathrm{d}Q}{\mathrm{d}\Omega} = \frac{4(2L'+1)(2S'+1)}{k_p^3 k_q} \sum_{\substack{\lambda J \\ l l' \\ j j'}} (-1)^{1/2(l+j'-l'-j)}(2\lambda+1)(2J+1) f_\lambda(j j' l l' J)$$
$$\times T(l l' J) T(j j' J) P_\lambda(\hat{\boldsymbol{k}}_p \cdot \hat{\boldsymbol{k}}_q) \tag{14}$$

where $P_\lambda(x)$ is a Legendre polynomial. The total cross section is given by

$$Q = \frac{16\pi(2L'+1)(2S'+1)}{k_p^3 k_q} \sum_{l l' J} (2J+1) T(l l' J)^2. \tag{15}$$

For the situation in which $L = L' = 0$, $J = 0$, and $l = l'$ equation (14) can be expressed in the form

$$\frac{\mathrm{d}Q}{\mathrm{d}\Omega} = \frac{4(2S'+1)}{k_p^3 k_q} \left(\sum_l (2l+1)^{1/2} T(l, l, 0) P_l(-\hat{\boldsymbol{k}}_p \cdot \hat{\boldsymbol{k}}_q) \right)^2. \tag{16}$$

3. Results

We first consider the excitation of the 2^3S state from the ground state for which there exists a substantial amount of experimental data. Our calculated total cross sections are shown in table 1 and some differential cross sections in table 2.

In figure 1 our calculated total cross section for excitation of the 2^3S is displayed as a function of (E/E_T) where in the figures E denotes the energy of the incident electron and

Table 1. Computed total cross sections for excitation of the 2^3S, 3^3S and 4^3S states (in units of πa_0^2). A superscript indicates the power of ten by which the number should be multiplied

Incident electron energy (eV)	$Q(1^1S–2^3S)$	$Q(1^1S–3^3S)$	$Q(1^1S–4^3S)$
20·4	$3{\cdot}68^{-2}$		
21·76	$5{\cdot}81^{-2}$		
23·12	$6{\cdot}29^{-2}$		
24·48	$6{\cdot}19^{-2}$	$1{\cdot}32^{-2}$	$3{\cdot}86^{-3}$
27·21	$5{\cdot}36^{-2}$	$1{\cdot}44^{-2}$	$5{\cdot}47^{-3}$
29·6	$4{\cdot}52^{-2}$	$1{\cdot}29^{-2}$	$5{\cdot}21^{-3}$
40·1	$2{\cdot}14^{-2}$	$6{\cdot}61^{-3}$	$2{\cdot}71^{-3}$
50	$1{\cdot}25^{-2}$	$3{\cdot}77^{-3}$	$1{\cdot}65^{-3}$
82	$4{\cdot}24^{-3}$	$1{\cdot}44^{-3}$	$5{\cdot}61^{-4}$
100	$2{\cdot}74^{-3}$	$8{\cdot}33^{-4}$	$3{\cdot}66^{-4}$
200	$5{\cdot}14^{-4}$	$1{\cdot}55^{-4}$	$6{\cdot}78^{-5}$

Table 2. Computed differential cross sections for excitation of the 2^3S state ($a_0^2\,sr^{-1}$). A superscript indicates the power of ten by which the number should be multiplied

θ (degrees)	E(eV)				
	29·6	40·1	82	100	200
0	$1{\cdot}08^{-2}$	$2{\cdot}49^{-2}$	$1{\cdot}96^{-2}$	$1{\cdot}53^{-2}$	$5{\cdot}27^{-3}$
10	$9{\cdot}47^{-3}$	$2{\cdot}17^{-2}$	$1{\cdot}56^{-2}$	$1{\cdot}17^{-2}$	$3{\cdot}19^{-3}$
20	$6{\cdot}12^{-3}$	$1{\cdot}43^{-2}$	$8{\cdot}25^{-3}$	$5{\cdot}63^{-3}$	$1{\cdot}01^{-3}$
30	$2{\cdot}57^{-3}$	$6{\cdot}89^{-3}$	$3{\cdot}19^{-3}$	$2{\cdot}00^{-3}$	$2{\cdot}75^{-4}$
40	$3{\cdot}70^{-4}$	$2{\cdot}07^{-3}$	$9{\cdot}59^{-4}$	$5{\cdot}79^{-4}$	$7{\cdot}32^{-5}$
50	$1{\cdot}50^{-4}$	$1{\cdot}53^{-4}$	$1{\cdot}88^{-4}$	$1{\cdot}18^{-4}$	$1{\cdot}37^{-5}$
55	$7{\cdot}35^{-4}$	$3{\cdot}72^{-6}$	$5{\cdot}47^{-5}$	$3{\cdot}79^{-5}$	$3{\cdot}70^{-6}$
60	$1{\cdot}69^{-3}$	$2{\cdot}14^{-4}$	$4{\cdot}66^{-6}$	$4{\cdot}90^{-6}$	$1{\cdot}86^{-7}$
70	$4{\cdot}38^{-3}$	$1{\cdot}27^{-3}$	$3{\cdot}55^{-5}$	$1{\cdot}57^{-5}$	$3{\cdot}61^{-6}$
80	$7{\cdot}59^{-3}$	$2{\cdot}66^{-3}$	$1{\cdot}43^{-4}$	$7{\cdot}39^{-5}$	$1{\cdot}29^{-5}$
90	$1{\cdot}08^{-2}$	$4{\cdot}07^{-3}$	$2{\cdot}75^{-4}$	$1{\cdot}48^{-4}$	$2{\cdot}34^{-5}$
100	$1{\cdot}39^{-2}$	$5{\cdot}34^{-3}$	$4{\cdot}10^{-4}$	$2{\cdot}25^{-4}$	$3{\cdot}32^{-5}$
110	$1{\cdot}66^{-2}$	$6{\cdot}44^{-3}$	$5{\cdot}35^{-4}$	$2{\cdot}97^{-4}$	$4{\cdot}16^{-5}$
120	$1{\cdot}88^{-2}$	$7{\cdot}37^{-3}$	$6{\cdot}46^{-4}$	$3{\cdot}60^{-4}$	$4{\cdot}85^{-5}$
130	$2{\cdot}07^{-2}$	$8{\cdot}13^{-3}$	$7{\cdot}42^{-4}$	$4{\cdot}14^{-4}$	$5{\cdot}41^{-5}$
140	$2{\cdot}22^{-2}$	$8{\cdot}73^{-3}$	$8{\cdot}19^{-4}$	$4{\cdot}57^{-4}$	$5{\cdot}84^{-5}$
150	$2{\cdot}33^{-2}$	$9{\cdot}19^{-3}$	$8{\cdot}79^{-4}$	$4{\cdot}91^{-4}$	$6{\cdot}16^{-5}$
160	$2{\cdot}41^{-2}$	$9{\cdot}51^{-3}$	$9{\cdot}22^{-4}$	$5{\cdot}14^{-4}$	$6{\cdot}37^{-5}$
170	$2{\cdot}45^{-2}$	$9{\cdot}70^{-3}$	$9{\cdot}47^{-4}$	$5{\cdot}29^{-4}$	$6{\cdot}51^{-5}$
180	$2{\cdot}47^{-2}$	$9{\cdot}76^{-3}$	$9{\cdot}55^{-4}$	$5{\cdot}33^{-4}$	$6{\cdot}54^{-5}$

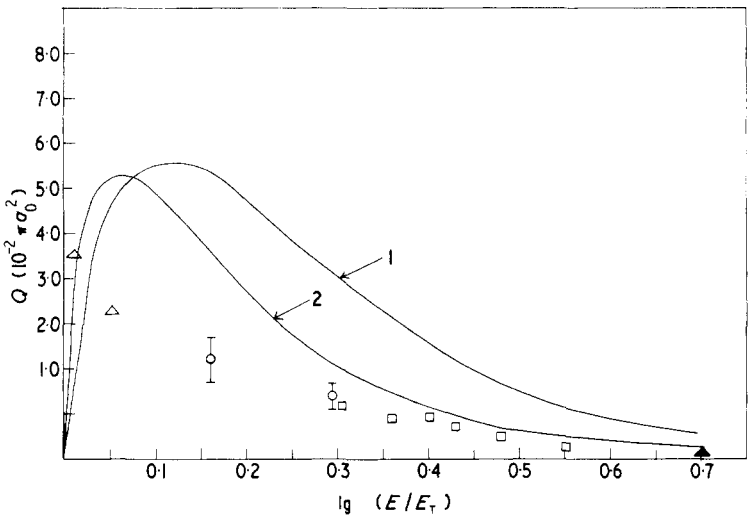

Figure 1. Total cross section for excitation of the 2^3S state. 1. Modified Ochkur approximation (Morrison and Rudge 1967), 2. present results, △ Brongersma *et al* 1972, ⏀ Trajmar 1973, □ Crooks *et al* 1972, ▲ Vriens *et al* 1968.

E_T is the threshold energy for the excitation process. Shown also in the figure are the results of four different experimental measurements together with the best previous high energy calculation, that of Morrison and Rudge (1967) who used the modified Ochkur approximation. It can be seen that our results are, for most energies, about a factor of

two smaller than the results of Morrison and Rudge (1967) which in themselves are smaller and therefore in better accord with experiment than several other theoretical methods which are not shown in the figure (cf Rudge 1973). Our results above about 50 eV are in reasonably good agreement with experimental data and below this energy are good to within about a factor of two. We cannot obtain with this technique the resonance structure near threshold which is not depicted in figure one nor do we obtain the structure reported by Crooks *et al* (1972) at 45 eV.

In order to assess the results more closely we examine the differential cross sections which are illustrated in figures 2 to 5.

Figure 2 compares our results at the low incident energy of 29·6 eV with the experimental measurements of Trajmar (1973) and with the results of the modified Ochkur and Oppenheimer methods quoted by Trajmar (1973). It can be seen from the figure that the modified Ochkur approximation is much better than the Oppenheimer approximation, which is in error by a factor of one hundred, but that, as noted by Vriens *et al* (1968) its shape is wrong in that the experimental measurements indicate a peak at 0° while the Ochkur approximation does not. Our approximation, which at this energy gives a total cross section which is close to that of the modified Ochkur method has a quite different shape. It does peak at 0° and decreases to a minimum near 50°. These features are in harmony with the experimental result though the calculated minimum is smaller. This is partially though perhaps not wholly due to the finite ($\approx 3°$) resolution of the experiment. The main defect of our method at this energy however clearly lies in our overestimate of the large angle scattering which in turn leads to an overestimate of the total cross sections.

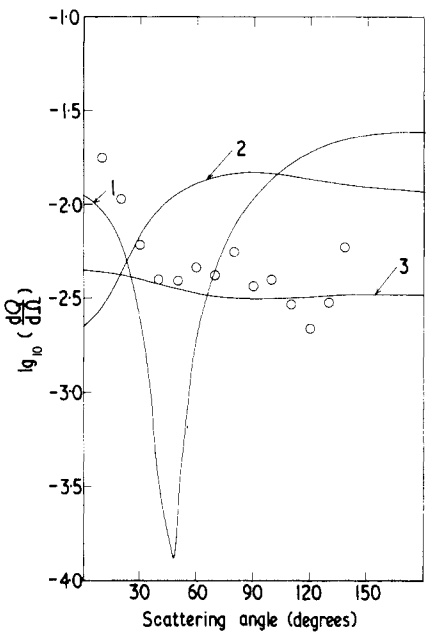

Figure 2. The differential cross section for excitation of the 2^3S state, $E_i = 29{\cdot}6$ eV. 1. Present results, 2. modified Ochkur results (Trajmar 1973), 3. $10^{-2}\times$ Born–Oppenheimer results (Trajmar 1973), ○ experimental measurements (Trajmar 1973).

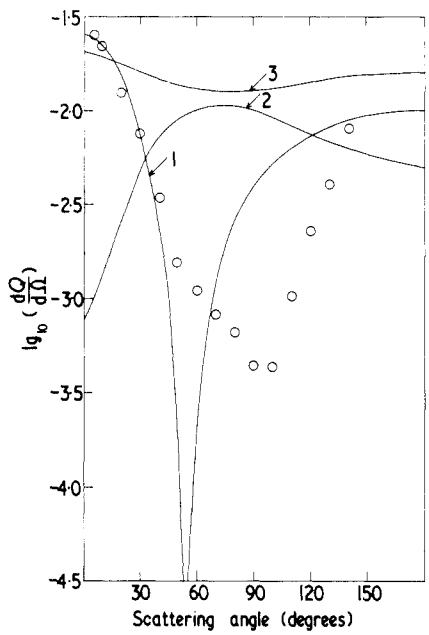

Figure 3. The differential cross section for excitation of the 2^3S state, $E_i = 40{\cdot}1$ eV. 1. Present results, 2. modified Ochkur results (Trajmar 1973), 3. $10^{-1}\times$ Born–Oppenheimer results (Trajmar 1973), ○ experimental measurements (Trajmar 1973).

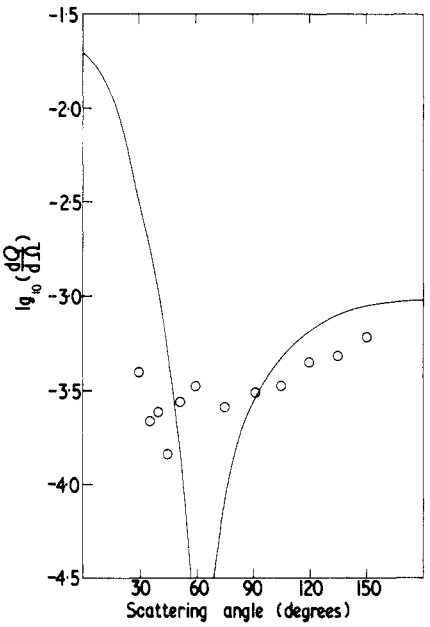

Figure 4. The differential cross section for excitation of the 2^3S state, $E_i = 82$ eV. ○ experimental results of Opal and Beaty 1972.

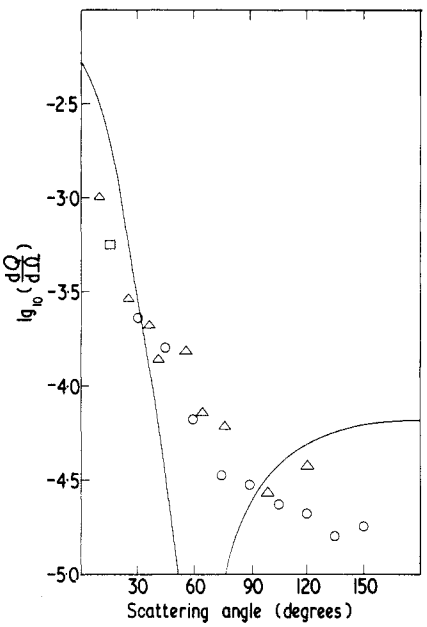

Figure 5. The differential cross section for excitation of the 2^3S state, $E_i = 200$ eV. ○ experimental results of Opal and Beaty 1972, △ experimental results of Suzuki and Takayanagi 1973, □ experimental results of Vriens *et al* 1968.

Figure 3 shows the results which we obtain at 40·1 eV. At scattering angles less than 30° we find excellent agreement with the results of Trajmar (1973). The calculated large angle scattering is relatively much less at this energy but we do not obtain the correct shape between 60° at 120°. The results however represent a very substantial improvement over the other theoretical treatments which also make use of plane waves.

The results at 82 eV, depicted in figure 4, illustrate the pattern that, as the energy increases so the cross section peaks more at small angles while there is correspondingly less scattering near 180°. Above 90° our results compare favourably with the measurements of Opal and Beaty (1972). The pronounced dip which we find near 60° is absent in the experimental results however and the disparity at 30° is surprisingly large.

Figure 5 illustrates the cross section at an energy of 200 eV. There is some disparity between the experimental data at large scattering angles: at small scattering angles, which at this energy contributes most strongly to the total cross section, there is reasonable accord with the experimental data. Near 60° however, as at the lower energies, we obtain a strong dip in the differential cross sections which is not reflected in the experimental results.

There exists less data for excitation of the n^3S states with $n > 2$. Figure 6 shows our calculated total cross section for excitation of the 3^3S state together with the experimental values of St John *et al* (1964). The cross section is sensitive to the choice of bound state 3^3S function and we do not therefore show other theoretical results from which it would be difficult to separate those differences which obtain from the bound state approximation and thqse which derive from the scattering approximation. Figure 7 shows the differential cross section at two energies for excitation of the 3^3S state. There is a clear similarity between these and the corresponding 2^3S results.

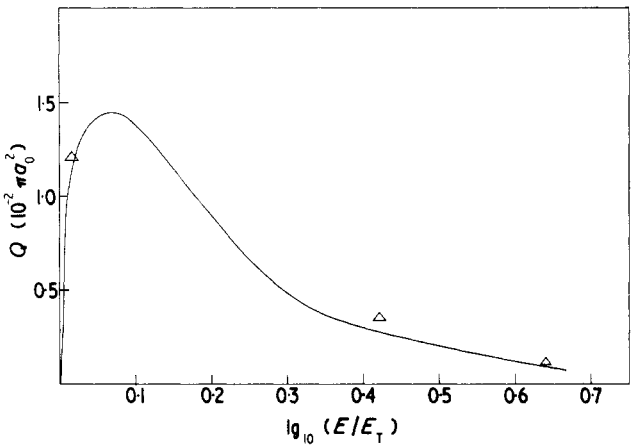

Figure 6. The total cross section for excitation of the 3^3S state. Δ Experimental data of St John *et al* 1964.

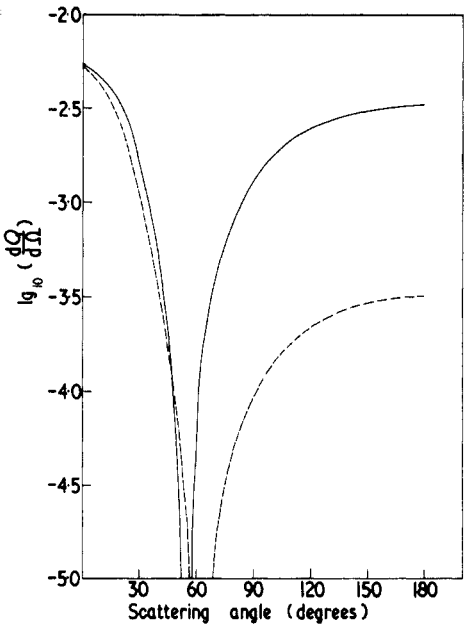

Figure 7. Calculated differential cross section for excitation of the 3^3S state at 40·1 eV (———) and 82 eV (– – –).

Figure 8 depicts the 4^3S total cross section. The relative experimental data of Kay and Showalter (1971) and of Anderson *et al* (1973) has been normalized to the result of St John *et al* (1964) at 100 eV. There is a large discrepancy between the resulting shape of the experimental cross section curves. Our results are close to those of St John *et al* (1964). Figure 9 illustrates the differential cross sections at two energies again showing close similarity with the 2^3S results.

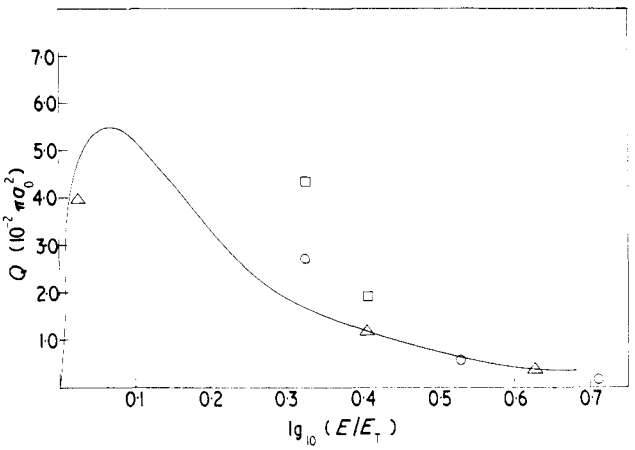

Figure 8. The total cross section for excitation of the 4^3S state: △ Experimental data of St John *et al* (1964), □ experimental data of Kay and Showalter (1971), ○ experimental data of Anderson *et al* (1973).

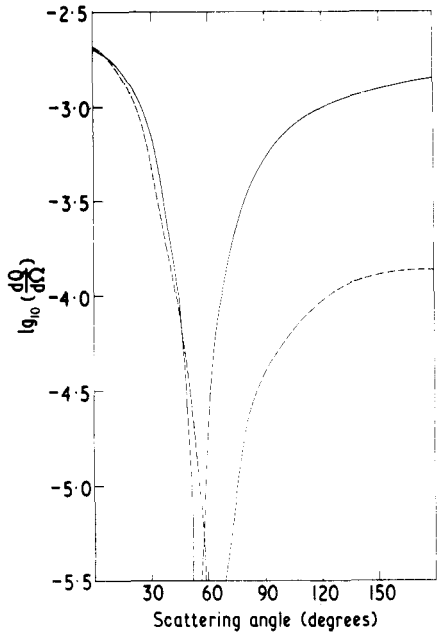

Figure 9. Calculated differential cross sections for excitation of the 4^3S state at 40·1 eV (———) and at 82 eV (– – –).

4. Conclusions

Our results demonstrate that our method is greatly superior to the Oppenheimer approximation, which is in error by one or two orders of magnitude for these cross sections, and superior also to the Ochkur approximation either in its modified or unmodified form. This is particularly evident where the differential cross sections are

concerned. These differential cross sections are extremely sensitive to the individual $T(l, l', J)$ values and therefore constitute a severe test of any theory. While therefore the agreement between our theoretical results and experiment is far from perfect, it is nonetheless encouraging that we do produce some of the salient features of the experimental measurements. We conclude that our procedure enables pure exchange cross sections to be computed to a similar degree of accuracy as are non-exchange cross sections by means of Born's approximation.

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