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LETTER TO THE EDITOR

A quantal study of differential cross sections for double charge transfer in C^{4+}–He collisions

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Abstract. Differential cross sections for the double charge transfer process in C^{4+}-He collisions are studied using a quantal two-channel molecular orbital close coupling expansion method. The calculated results for C^{4+} at incident energies of 1520 and 500 eV are found to be in good agreement with recent measurements. It is also shown that the quantal calculations and the experimental results are well reproduced by a semiclassical approximation. Comparison with the recent semi-empirical study of the same system by Barany *et al* is also discussed.

The charge transfer process between multiply charged ions and neutral atoms has been the subject of much theoretical and experimental investigation in recent years (Janev and Winter 1985). Most of the experiments carried out with different ion sources and with recoil ions provide total charge transfer cross sections. In the last few years, with the introduction of energy-gain spectroscopy (Schmeissner *et al* 1984, Okuno *et al* 1983) and with optical measurements (Dijkkamp *et al* 1985), charge transfer to specific final states has been reported. Only in the last two years, experimental differential cross sections for charge transfer processes from different ion sources have been carried out at several laboratories.

In this letter we report a theoretical study on the differential cross section for the double charge transfer process in C^{4+}-He collisions. For this system there are measurements from two laboratories at several energies using recoil ion sources (Cederquist *et al* 1985, Barany *et al* 1986). In the low-energy region (500–1500 eV for C^{4+}) studied here, double electron capture of He by C^{4+} ($1s^2$) to C^{2+} ($1s^2\,2s^2$) is the dominant process and thus the collision can be treated as a two-channel problem. We adopt a full quantum mechanical formulation; thus the motions of the electrons and of the heavy particles are both treated quantum mechanically. The results are also analysed to check the validity of the semiclassical approximations.

This collision system has also been studied theoretically by Barany *et al* (1986). These authors adopted a semiclassical two-channel approximation. They fitted the relevant potential curves and coupling terms so that experimental differential cross sections are well reproduced. Our approach is different in that the relevant potential curves and the coupling terms are obtained through *ab initio* calculations and the differential cross sections are calculated in a full quantal formulation. Our goal is to show that the present *ab initio* calculations are capable of reproducing these differential cross sections. We further show that in this energy range, a semiclassical calculation is also valid.

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In a full quantum formulation of heavy particle scattering at low energies (Heil *et al* 1981), the total Hamiltonian includes two parts—an electronic part and a heavy-particle part. The total wavefunction of the system in the adiabatic representation is given by

$$\Psi(\boldsymbol{R}, \boldsymbol{r}) = \sum_n F_n^a(\boldsymbol{R}) \chi_n^a(R; \boldsymbol{r}) \tag{1}$$

where $\boldsymbol{R}$ is the internuclear separation and $\boldsymbol{r}$ denotes the electronic coordinates. In (1), $F_n(\boldsymbol{R})$ is the wavefunction of the heavy particle when the electrons are in state n and the $\chi_n(R; \boldsymbol{r})$ are the adiabatic molecular wavefunctions including electron translational factors (ETF). It is more convenient to solve differential equations for heavy-particle wavefunctions in a diabatic representation. Transformation from adiabatic to diabatic representation is achieved through a transformation matrix $C(R)$. In this representation the radial wavefunction for the heavy particles is $F^d = \mathbf{C}^{-1} F^a$, and the diabatic potential matrix is $\mathbf{V}^d = \mathbf{C}^{-1} \mathbf{V}^a \mathbf{C}$, where $\mathbf{V}^a$ is the adiabatic potential matrix.

In the diabatic representation F^d satisfies (in atomic units)

$$[(-1/2\mu)\nabla_R^2 \mathbf{I} - \mathbf{V}^d(R) + E\mathbf{I}] F^d(\boldsymbol{R}) = 0 \tag{2}$$

where $\mathbf{I}$ is the identity matrix, and μ is the reduced mass. Equation (2) becomes a coupled radial equation by a partial wave expansion of $F^d(\boldsymbol{R})$. The resulting radial equation for each partial wave is then solved numerically using the log-derivative method of Johnson (1973). From the S matrix calculated for each partial wave, the differential cross section is obtained from the standard formula

$$\frac{d\sigma}{d\Omega} = \frac{1}{2k^2} \left| \sum_l (2l+1) S_{12}^l P_l(\cos\theta) \right|^2 \tag{3}$$

where S_{12}^l is the off-diagonal matrix element for partial wave l, k is the momentum of the incident particle in the centre of mass frame and θ is the angle of the scattered particle with respect to the incident direction.

We have carried out a two-channel full quantum calculation for C^{4+} on He to obtain the differential charge transfer cross section at impact energies 1520 and 500 eV (13.97 and 4.60 au in centre of mass). The adiabatic potential curves (matrix $\mathbf{V}^a$) and coupling term used for this work are taken from Kimura and Olson (1984). The readers are referred to that article for the relevant potential curves. The boundary condition for the transformation matrix is $\mathbf{C}(R \to \infty) = \mathbf{I}$. In figure 1 we show the results of the theoretical calculation and its comparison with the measurement of Cocke *et al* (see Barany *et al* 1986). The theoretical calculation has been folded with an experimental angular resolution of 4.8 mrad (L Tunnell, private communication) and the relative experimental measurement has been normalised for comparison. We note that the experimental oscillatory structure is well reproduced by the theoretical calculation, particularly the positions of maxima and minima and their relative heights. On the other hand, there is a small but noticeable shift in the position of the first maximum.

A similar comparison with the experimental data of Cederquist *et al* (1985) for collision energy at 500 eV is shown in figure 2. Here the differential cross section is displayed against the energy gain of the projectile. The theoretical results are convoluted with an energy resolution of 1.2 eV (J Pedersen, private communication). It can be seen that the theoretical energy-gain spectra are in reasonable agreement with the data. It is noted that if the experimental data are shifted by 1 eV so that its highest peak coincides with the theoretical result, the positions of the other pronounced maxima

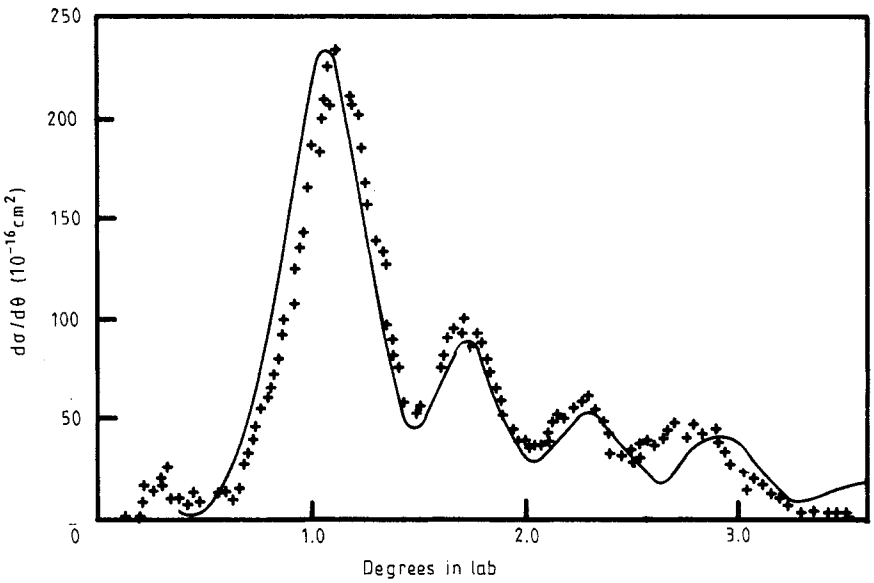

Figure 1. Differential charge transfer cross section for C^{4+} collision on He at 1520 eV (the relative velocity $V = 0.071$ au). Experimental data are from Tunnell and Cocke (Barany *et al* 1986). The theoretical cross sections are given in units of 10^{-16} cm^2 and convoluted with experimental resolution (full line). Experimental data are in abitrary units.

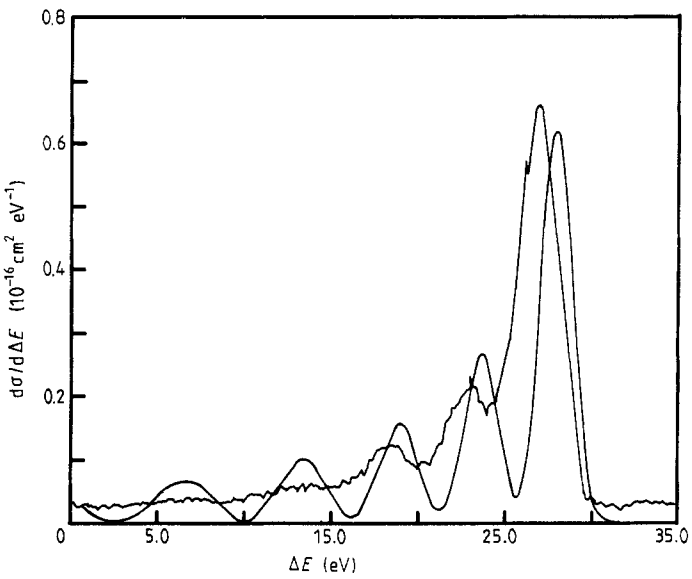

Figure 2. Energy-gain spectrum for double charge transfer process for C^{4+} on He at 500 eV (the relative velocity $V = 0.041$ au). Experimental data: Cederquist *et al* (1985), arbitrary units. Theoretical result: full line in units of 10^{-16} cm^2 eV^{-1}.

and minima would then be in good agreement with the theoretical calculations. In obtaining the energy-gain spectrum from the calculated angular distribution, we use the relation

$$Q = 2\gamma[E_0(E_0+\Delta E)]^{1/2}\cos\theta + (1+\gamma)(E_0+\Delta E) + (1-\gamma)E_0 \qquad (4)$$

where E_0 is the incident energy, ΔE is the energy gain, Q is the energy defect between initial and final states and γ is the ratio of the mass of C to that of He.

The total double charge transfer cross sections at 500 and 1520 eV were calculated to be 4.079×10^{-16} cm^2 and 4.098×10^{-16} cm^2, respectively. These results are larger than those obtained from a multichannel calculation done by Kimura and Olson (1984). This discrepancy can be attributed to the idealised two-channel model used in the present calculation which tends to overestimate total cross sections. The numbers of partial waves needed to reach a reasonable convergence for the total cross section calculation for 1520 and 500 eV are about 1500 and 900, respectively.

The results in figures 1 and 2 indicate that the experimental differential charge transfer cross sections are well described by a two-channel full quantal model. It is desirable to see if the major features of these differential cross sections can be explained by semiclassical methods. To this end, we first show in figure 3 (full curve) $|S_{12}^l|^2$ at 1520 eV obtained from the quantal calculation against total angular momentum. The broken curves in the figure are from the semiclassical calculation which will be discussed later. Figure 3 shows that the magnitude of S_{12} is oscillatory. The irregularity in the oscillation at small l is due to the inaccuracy of the *ab initio* potential curves in the small-R region. Since the potential curves were calculated using a pseudopotential for C^{4+}, the potential is not expected to be valid at small R. However, this irregularity at small l would not affect the results of this study since their contribution to the measured angular region is very minimal.

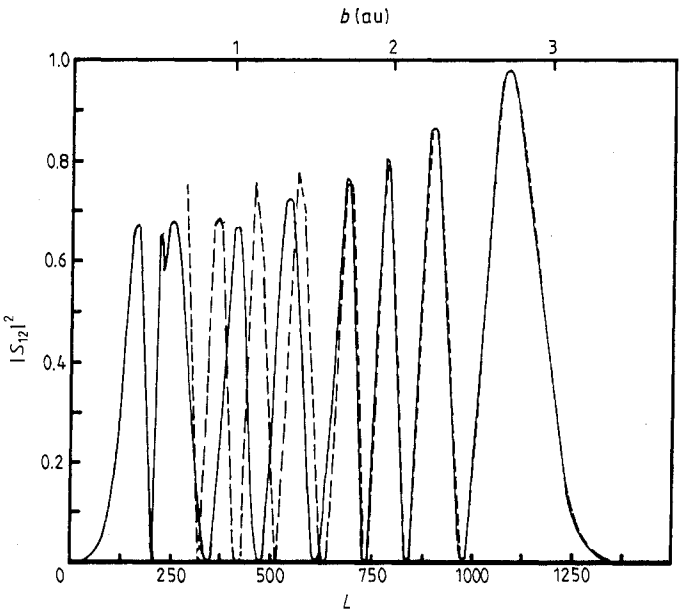

Figure 3. $|S_{12}|^2$ against partial waves at impact energy 1520 eV. The full curve is the result of a full quantal calculation. The broken curve is from a semiclassical calculation using a curve trajectory (see text).

We next consider how the oscillatory features of the differential cross sections are related to the oscillatory structure of $|S_{12}|$. For this purpose S_{12}^l is expressed as

$$S_{12}(l) = A^l \sin \xi^l \exp(i\eta_{12}^l) \tag{5}$$

where A^l is a slowly varying function of l and the oscillation of $|S_{12}|$ is governed by the function $\sin \xi^l$. Note that, from (5), the scattering amplitude $f(\theta)$ is the sum of the product of three oscillatory functions, namely the two functions $\sin \xi^l$ and $\exp(i\eta_{12}^l)$ of (5) and the Legendre polynomial $P_l(\cos\theta)$ which is approximately given by

$$P_l(\cos\theta) = \left(\frac{2}{(l+\frac{1}{2})\pi \sin\theta}\right)^{1/2} \sin[(l+\tfrac{1}{2})\theta + \tfrac{1}{4}\pi] \tag{6}$$

for $l \sin\theta \gg 1$. This oscillatory sum can be evaluated using a stationary phase approximation (Mott and Massey 1949). Constructive interference occurs only near the l value, $l\pm = l_\theta$, where the phase is stationary, i.e. where the first derivative of the phase with respect to l vanishes:

$$\theta_\pm = \frac{d}{dl}(\eta_{12}^l \pm \xi^l). \tag{7}$$

Thus $f(\theta)$ is evaluated at the two stationary phases, $l\pm = l_{\theta_\pm}$,

$$f(\theta) = C_+ e^{i\beta_+} + C_- e^{i\beta_-} \tag{8}$$

and

$$\frac{d\sigma}{d\Omega} = \frac{k_2}{k_1}[|C_+|^2 + |C_-|^2 + 2C_+^* C_- \cos(\beta_+ - \beta_-)] \tag{9}$$

where

$$C_\pm(\theta) = \frac{A_\pm}{2i\sqrt{k_1 k_2}}\left(\frac{l_\pm + \frac{1}{2}}{\sin\theta\,|d\theta/dl|_{l_\pm}}\right)^{1/2} \tag{10}$$

$$\beta_\pm(\theta) = (\eta_{12}^l + \xi^l)_{l_\pm} - (l_\pm + \tfrac{1}{2})\theta - \tfrac{1}{4}\pi \tag{11}$$

and k_1 and k_2 are the momenta of the incident and exit channels, respectively. From (7) it is natural to introduce θ_+ and θ_- as the two classical deflection functions of the two charge transfer 'paths'. The oscillatory term, $\cos(\beta_+ - \beta_-)$, may be interpreted as the interference due to the two paths. In the present studied system, $d\xi^1/dl$ is very small over a large range of l so that we can define a single deflection function

$$\Theta = \frac{d\eta_{12}^l}{dl}. \tag{12}$$

In this limit, $\beta_+ - \beta_- = 2\xi^l$, $C_+ = C_-$, and (9) becomes

$$\frac{d\sigma}{d\Omega} = 4\frac{k_2}{k_1}|C_+(\theta)|^2 \sin^2 \xi(\theta). \tag{13}$$

Note that (13) is proportioal to $|S_{12}|^2$ at an angle defined by (12). Equations (12) and (13) allow us to perform a simple mapping from $|S_{12}|^2$ to $d\sigma/d\Omega$ under the mathematical stationary phase approximation. Such a mapping is displayed in figure 4. In this figure, three quantities from the quantal calculations are compared with the corresponding ones from the semiclassical calculations.

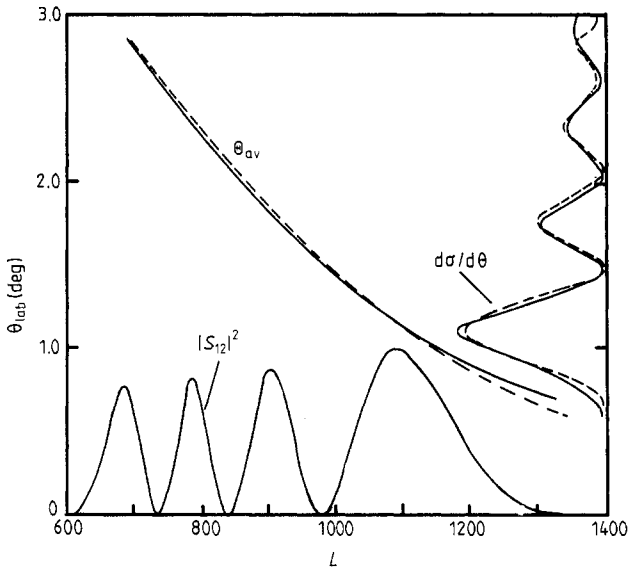

Figure 4. Mapping of electron capture probability to obtain differential cross sections via a deflection function. The full curves in the deflection function and in the differential cross sections are obtained from quantal calculations while the same quantities calculated by the semiclassical method are shown as broken curves.

(1) The quantal $|S_{12}|^2$ is displayed against classical impact parameter $b = L/k$. This quantity can be identified with the charge transfer probability $P(b)$ at a given impact parameter b calculated from the semiclassical model. Such a comparison is given in figure 3 where $P(b)$ is given by broken curves. The trajectory used in the semiclassical calculation is determined by an average diabatic potential, $V_{av} = \frac{1}{2}(V_{11}(R) + V_{22}(R))$. The agreement between the two calculations is quite good at large impact parameters (or large l). The discrepancy at small l does not affect the angular distribution at the small angles studied here.

(2) The deflection function calculated from (12), shown by full curves, is compared with the classical deflection function

$$\Theta(b) = \pi - 2b\sqrt{2mE}\int_{R_0}^{\infty}\frac{dR}{R^2 p(R)}. \tag{14}$$

In (14), R_0 is the turning point and $p(R)$ is the radial momentum calculated in the average potential V_{av}. The classical deflection function, as shown by broken curves, is not too different from the quantal one obtained from (12).

(3) The differential cross sections; using $|S_{12}|^2$ as the capture probability, together with the classical deflection function, one can obtain the differential cross sections by mapping from l to scattering angles. The results are shown by broken curves in figure 4. Notice that this simplified semiclassical result is in good agreement with the quantal calculation as well as with experimental data.

The agreement between the quantal and the semiclassical calculations deteriorates as the collision energy decreases. Generally, the number of oscillations in the semiclassical $P(b)$ is proportional to the inverse of the impact velocity while that of quantal $|S_{12}|^2$ is not. We have checked the comparison at 500 eV and the agreement between

quantal and semiclassial results is still reasonable. At lower energies the semiclassical method used here becomes invalid.

We have also used the empirically determined diabatic potentials V_{ij} of Barany *et al* (1986) to evaluate the differential cross sections in our quantal calculation. Although the resulting $d\sigma/d\theta$ are very similar to theirs (and in agreement with experiments), the details in S_{12} are actually quite different. The oscillation in $|S_{12}|^2$ from the deduced potentials is much more rapid than the $|S_{12}|^2$ shown in figure 3. It indicates that the rate of change in the phase of S_{12} is also larger in their calculation. It has been demonstrated by Dinterman and Delos (1977) by a semiclassical argument that the transition amplitude does not depend upon the three potential matrix elements, V_{ij}, taken independently, but only upon the behaviour of $(V_{11}-V_{22})/2V_{12}$. In other words, it is not possible to deduce potential curves uniquely from relative experimental differential cross sections for two-channel problems.

In summary, we have demonstrated in this letter that the measured differential charge transfer cross sections in $C^{4+}(1s^2)+He \rightarrow C^{2+}(1s^22s^2)+He^{2+}$ are well described by a model based on a quantal two-channel molecular orbital close-coupling expansion method. It is shown that the molecular expansion with the electron translational factors adopted in this calculation does give a good description of the measured differential cross sections. We have also illustrated that the present two-channel results can be described by a semiclassical theory. Recent measurements of differential cross sections involve systems which are inherently multichannel phenomena. It is desirable to pursue the question of whether these new data can be described by a full multichannel quantal calculation and if there is a corresponding semiclassical description.

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