Application of the eikonal amplitude to rotational excitations of diatomic molecules by electron impact

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The eikonal amplitude is used in the framework of the adiabatic approximation to obtain the rotational excitations of homonuclear diatomic molecules by intermediate-energy electron impact. The formalism is simple and may be extended to collision processes in which heteronuclear molecules are used as the target, or positrons as projectiles. The results for the differential cross section of the $J = 1 \rightarrow 3$ rotational excitation of hydrogen molecules by 40-eV electron impact are in qualitative agreement with the recent experimental findings.

Rotational excitations of diatomic molecules play a dominant role in the energy-loss mechanism for low-energy electron-molecule scattering processes and have been the subject of considerable theoretical and experimental investigations.[1] A recent experiment[2] in which hydrogen molecules were used as the target has revealed that rotational cross sections are not insignificant, particularly at large scattering angles, compared to pure elastic cross sections in the energy region 30–100 eV. It clearly indicates the importance of the theoretical study of rotational excitations of diatomic molecules by intermediate-energy electrons.

In this note we present a formulation for the initiation of this study using the eikonal amplitude.[3] Previously we have used the eikonal amplitude to investigate the average elastic (sum of pure elastic and rotational) cross sections of electrons[4] and positrons[5] scattered by molecular hydrogen. However, the formulation given in Ref. 4 cannot be applied directly to the investigation of pure rotational cross sections. As a test case, we calculate rotational differential cross sections for collisions of hydrogen molecules with 40-eV electrons and compare them with those obtained experimentally.[2]

In the intermediate-energy region the effective collision time is very much shorter than the period of molecular rotations, and the adiabatic approximation[1,6] is a valid description of the scattering process. In this approximation one needs, in order to calculate the rotational cross sections, only the amplitude of the elastic scattering of an electron from the target molecule held fixed in space. Here this elastic amplitude is considered in the eikonal approximation. Under the assumption that the target molecule is a rigid rotor, the differential cross section for the rotational excitation from J, M to J', M', in the adiabatic approximation, is given by[1] (atomic units are used throughout)

$$I(J, M \rightarrow J', M', \theta) = \left| \int Y^*_{J'M'}(\hat{R}) f(\theta, \hat{R}) Y_{JM}(\hat{R})\, d\hat{R} \right|^2. \tag{1}$$

The differential cross section for the excitation from J to J' is obtained by averaging over M and summing over M':

$$I(J \rightarrow J', \theta) = \frac{1}{2J+1} \sum_M \sum_{M'} \left| \int Y^*_{J'M'}(\hat{R}) f(\theta, \hat{R}) Y_{JM}(\hat{R})\, d\hat{R} \right|^2. \tag{2}$$

Taking the sum over J', M' and averaging over M in Eq. (1), we get the average elastic differential cross section,

$$\langle I(\theta) \rangle = \frac{1}{4\pi} \int |f(\theta, \hat{R})|^2\, d\hat{R}. \tag{3}$$

In these equations $\hat{R}$ is a unit vector along the internuclear separation $\vec{R}$, θ is the scattering angle, and the Y's are the spherical harmonics. The eikonal elastic scattering amplitude $f(\theta, \hat{R})$ is given by

$$f(\theta, \hat{R}) = -\frac{ik_i}{2\pi} \int e^{i\vec{q}\cdot\vec{b}_3} \times \left[\exp\left(-\frac{i}{v_i} \int_{-\infty}^{\infty} V(\vec{r}_3, \hat{R})\, dz_3 \right) - 1 \right] d^2 b_3, \tag{4}$$

where $m_e \vec{v}_i = \hbar \vec{k}_i$ is the momentum of the incident electron, and $\vec{q} = \vec{k}_i - \vec{k}_f$ is the momentum transfer to the target molecule. The position vector $\vec{r}_3$ of the electron is related to the impact parameter vector $\vec{b}_3$ by

$$\vec{r}_3 = \vec{b}_3 + \hat{k}_i z_3. \tag{5}$$

Here we have taken the center of gravity of the molecule as the origin, and the direction of $\vec{k}_i$ as the polar axis.

The electron-molecule interaction V can be expanded in terms of Legendre polynomials,

$$V(\vec{r}_3, \hat{R}) = \sum_\nu V^\nu(r_3) P_\nu(\hat{r}_3 \cdot \hat{R}), \tag{6}$$

with

$$\hat{r}_3 \cdot \hat{R} = \cos\theta_3 \cos\theta_m + \sin\theta_3 \sin\theta_m \cos(\phi_3 - \phi_m),$$

where θ_m, ϕ_m denote the orientation of $\vec{R}$ with respect to the polar axis. For homonuclear diatomic molecules the above summation contain 0 and even values of ν. We retain only first two terms, V^0 and V^2. Using Eqs. (6) and (5) in the expression for $f(\theta, \hat{R})$ [Eq. (4)] we get

$$f(\theta, \hat{R}) = -\frac{ik_i}{2\pi} \int e^{i\vec{q}\cdot\vec{b}_3} \{\exp[-i\alpha(b_3) + i\beta(b_3, \theta_m) - i\gamma(b_3, \theta_m) \cos 2(\phi_3 - \phi_m)] - 1\} d^2 b_3, \tag{7}$$

with

$$\alpha(b_3) = \frac{2}{v_i} \int_0^\infty \left(V^0(r_3) + \frac{V^2(r_3)}{2} \frac{2z_3^2 - b_3^2}{r_3^2} \right) dz_3,$$

$$\beta(b_3, \theta_m) = \frac{3}{2} \frac{\sin^2\theta_m}{v_i} \int_0^\infty V^2(r_3) \frac{2z_3^2 - b_3^2}{r_3^2} dz_3,$$

$$\gamma(b_3, \theta_m) = \frac{3}{2} \frac{\sin^2\theta_m}{v_i} \int_0^\infty V^2(r_3) \frac{b_3^2}{r_3^2} dz_3.$$

When the variable is changed from ϕ_3 to Φ, where $\Phi = \phi_3 - \phi_m$, Eq. (7) becomes

$$f(\theta, \hat{R}) = -\frac{ik_i}{2\pi} \left(\int \exp[iqb_3 \cos(\Phi + \phi_m) - i\gamma(b_3, \theta_m) \cos 2\Phi - i\chi(b_3, \theta_m)] b_3 \, db_3 \, d\Phi - 2\pi J_0(qb_3) b_3 \, db_3 \right), \tag{8}$$

where $\chi(b_3, \theta_m) = \alpha(b_3) - \beta(b_3, \theta_m)$. Expanding $\exp[iqb_3 \cos(\Phi + \phi_m) - i\gamma \cos 2\Phi]$ in terms of Bessel functions and carrying out the Φ integration, Eq. (8) reduces to

$$f(\theta, \hat{R}) = -ik_i \sum_{n=0} i^n \lambda_n f_{2n,n}(\theta, \theta_m) \cos 2n\phi_m, \tag{9}$$

with

$$f_{0,0}(\theta, \theta_m) = \int b_3 \, db_3 \{J_0(qb_3)[e^{-i\chi} J_0(\gamma) - 1]\},$$

$$f_{2n,n}(\theta, \theta_m) = \int b_3 \, db_3 \, e^{-i\chi} J_{2n}(qb_3) J_n(\gamma),$$

$$\lambda_n = \begin{cases} 1, & \text{for } n = 0, \\ 2, & \text{for } n \neq 0, \end{cases}$$

where the J_n's are the Bessel functions of order n.

Substituting Eq. (9) into (2) (and after some manipulations) we obtain

$$I(J \to J', \theta) = \frac{k_i^2(2J'+1)}{4} \sum_{n=0} \sum_{M=-J}^{J} \left| \left(\frac{(J - |M|)!}{(J + |M|)!} \frac{(J' - |M| + 2n)!}{(J' + |M| - 2n)!} \right)^{1/2} P_J^{|M|}(\cos\theta_m) f_{2n,n}(\theta, \theta_m) P_{J'}^{|M|-2n}(\cos\theta_m) \sin\theta_m \, d\theta_m \right|^2, \tag{10}$$

with the condition that $2n = |M - M'|$. The average elastic differential cross section is obtained using Eq. (9) in Eq. (3):

$$\langle I(\theta) \rangle = \frac{k_i^2}{4\pi} \int \left(\sum_{n=0} \lambda_n^2 f_{2n,n}^2(\theta, \theta_m) \cos^2 2n\phi_m + \sum_{\substack{n,p=0 \\ n \neq p \\ n+p \text{ even}}} \lambda_n \lambda_p i^{3p+n} f_{2n,n}(\theta, \theta_m) f_{2p,p}(\theta, \theta_m) \cos 2n\phi_m \cos 2p\phi_m \right) d\hat{R}. \tag{11}$$

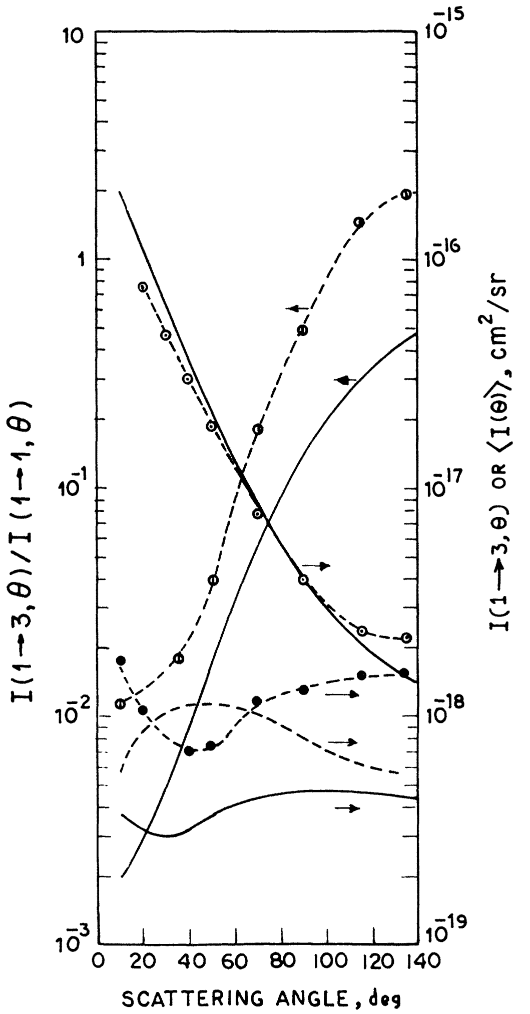

FIG. 1. Left-hand-side ordinates, ratio $I(1\to 3,\ \theta)/I(1\to 1,\ \theta)$: ⦶, Ref. 2; —, present calculations. Right-hand-side ordinates, average differential cross sections $\langle I(\theta)\rangle$: ⊙, Ref. 2; —, present calculations; pure rotational excitation cross sections $I(1\to 3,\ \theta)$: ●, Ref. 2; – – –, theoretical calculations at 45 eV as reported in Ref. 2; —, present calculations. (Arrows show which scales apply.)

The cross terms containing cosines of ϕ_m vanish with the ϕ_m integration, giving

$$\langle I(\theta)\rangle = \frac{k_i^2}{2}\sum_{n=0}\int \lambda_n f^2_{2n,n}(\theta,\theta_m)\sin\theta_m\, d\theta_m. \qquad (12)$$

The expression (10) and a comparison of Eq. (13) of Ref. 4 with Eq. (12) above reveal that the present formulation is more general and elegant than that reported earlier[4,5] in that (i) we can now compute pure elastic rotational as well as average elastic cross sections separately, (ii) to compute average elastic cross sections we are no longer required to take the average of three chosen molecular orientations,[4,5] which is questionable at large scattering angles and for molecules having strong nonspherical potentials, and (iii) the physical significance of the rapid convergence of the terms $f_{2n,n}$ involving higher-order Bessel functions in the eikonal amplitude can be well understood (higher-order Bessel functions contribute only to transitions involving higher rotational levels). In addition, the present method consumes less computer time.

Finally, we calculate the average elastic cross sections [Eq. (12)], rotational differential cross section [Eq. (10)], and the ratio $I(1\to 3)/I(1\to 1)$ for the e^--H_2 system at 40 eV. We use the static potentials V_s^0 and V_s^2 of Ref. 4 and the long-range potentials of Ref. 5 (model A). The effect of exchange is not considered. The results are shown in Fig. 1 and are compared with the corresponding experimental quantities of Ref. 2 and 7.

Our results for the average elastic cross section are found to be in good agreement with the observed values in the angular region 20°–120°. We obtained similar agreement earlier.[4] The rotational excitation ($1\to 3$) cross section gives qualitative agreement with the experimental observation. We obtain a minimum in the rotational cross section at 35°, whereas the observed minimum is at 40°. The calculated cross sections are, on the average, 2.5 times less in magnitude than the observed values. For the sake of comparison we have shown in Fig. 1 the calculated $I(1\to 3)$ at 45 eV, as reported in Ref. 2. This theoretical curve of Ref. 2 fails to reproduce the nature of the observed values. Unlike the experimental observation the ratio $I(1\to 3)/I(1\to 1)$ never exceeds unity even at large angles. The discrepancies between the present results and the observed values may be minimized if the effect of exchange is taken properly into account.

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