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Multiple differential cross sections for single ionization of H_2, D_2, and T_2 molecules by fast electron impact: Influence of vibrational states

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Multiple differential cross sections for $(e,2e)$ reactions are computed for H_2, D_2, and T_2 targets at high-impact energies by using a first-order Born approximation within the formal theory. Results are compared with previously existing calculations. The influence of vibrational states in obtaining differential cross sections is determined for an asymmetric coplanar geometry.

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One of the most important processes in atomic physics is ionization of atomic and molecular targets by electron impact. In particular, this reaction is of interest in many fields such as plasma physics, controlled fusion devices, and radiobiology. In previous works, the $(e,2e)$ process for simple molecular targets was studied (see, for instance, Refs. [1–5]). The present work deals with single ionization of diatomic molecules of hydrogen, deuterium, and tritium by impact of fast electrons. Recently (see Ref. [6], hereafter referred to as I), transitions between the lowest vibrational level of electronic ground state ${}^1\Sigma_g^+$ of H_2, D_2, and T_2, to final vibrational states of the ${}^2\Sigma_g^+$ electronic states of $H_2{}^+$, $D_2{}^+$, and $T_2{}^+$, were studied theoretically by calculating the fivefold differential cross section (5DCS) of this process by a model in which the electronic transition probability is modulated by the probability density of the initial and final vibrational levels. It was assumed, as usual for high-impact-energy values, that the ionization reaction takes place through vertical transitions, i.e., at fixed molecular internuclear distances. Moreover, the molecular internuclear vector $\boldsymbol{\rho}$ was supposed to remain frozen during the collision.

Multiply differential cross sections were thus expressed as the product of the electronic transition matrix element $t_{fi}^e(\boldsymbol{\rho})$ and the probability density of finding the diatomic target at a given internuclear distance. The sevenfold cross section was obtained by employing the expression

$$\sigma^{(7)}_{\nu_f,\nu_i}=\frac{d^7\sigma}{d\Omega_\rho\, d\Omega_e\, d\Omega_s\, d(k_s^2/2)} =(2\pi)^4\frac{k_e\, k_s}{k_i}\int_0^\infty d\rho\,\rho^2|\Xi_{\nu_f}(\rho)|^2|\Theta_{\nu_i}(\rho)|^2|t_{if}^e(\boldsymbol{\rho})|^2 \tag{1}$$

The momenta of the incident, scattered, and ionized electrons are indicated by k_i, k_s, and k_e, respectively. The solid angles corresponding to $\boldsymbol{\rho}$, k_e and k_s are denoted by Ω_ρ, Ω_e, and Ω_s, respectively. $|\Theta_{\nu_i}(\rho)|^2$ and $|\Xi_{\nu_f}(\rho)|^2$ represent the probability densities of the initial and final vibrational states for the lowest rotational levels, respectively. Neglecting the exchange and capture terms, the transition matrix element reduces to twice the direct term[1] (see Ref. [7] for a detailed explanation). Averaging over all molecular orientations one obtains the 5DCS as

$$\sigma^{(5)}_{\nu_f,\nu_i}=\frac{1}{4\pi}\int d\Omega_\rho\sigma^{(7)}_{\nu_f,\nu_i}. \tag{2}$$

These cross sections exhibited a strong dependence on the final vibrational states. Making the summation over all possible final vibrational states, important differences between the 5DCS's of H_2, D_2, and T_2 were obtained for an energy resolution of 3 eV which involves transitions from the initial rotational, vibrational, and electronic ground states of the target to all the vibrational levels of the ground electronic state of the corresponding residual ions $H_2{}^+$, $D_2{}^+$, and $T_2{}^+$.

In this work we repeat the same calculations by employing the formal theory in which the vibrational and rotational wave functions are introduced (in contrast with I) in the first-order Born approximation of the transition matrix element. Now, using the adiabatic discription of the molecules, we write the initial and final eigenfunctions as a product of electronic, vibrational, and rotational wave functions as

$$\psi_i=\psi_{e_i}(\mathbf{q},\boldsymbol{\rho})\Theta_{\nu_i J}(\rho)Y_{JM}(\Omega'_\rho) \tag{3}$$

and

$$\psi_f=\psi_{e_f}(\mathbf{q},\boldsymbol{\rho})\Xi_{\nu_f J'}(\rho)Y_{J'M'}(\Omega'_\rho), \tag{4}$$

where M and M' are the eigenvalues of the rotational angular momentum $\mathbf{J_z}$ defined with respect to a body fixed frame, and $\mathbf{q}$ denotes all the electronic coordinates.

Let us consider the sum of the 5DCS's corresponding to the transitions from a given initial state (ν,J,M) of the ground electronic state of the target to all the rotational

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[1] There is a misprint in I. The multiplicative factor 2 is missing.

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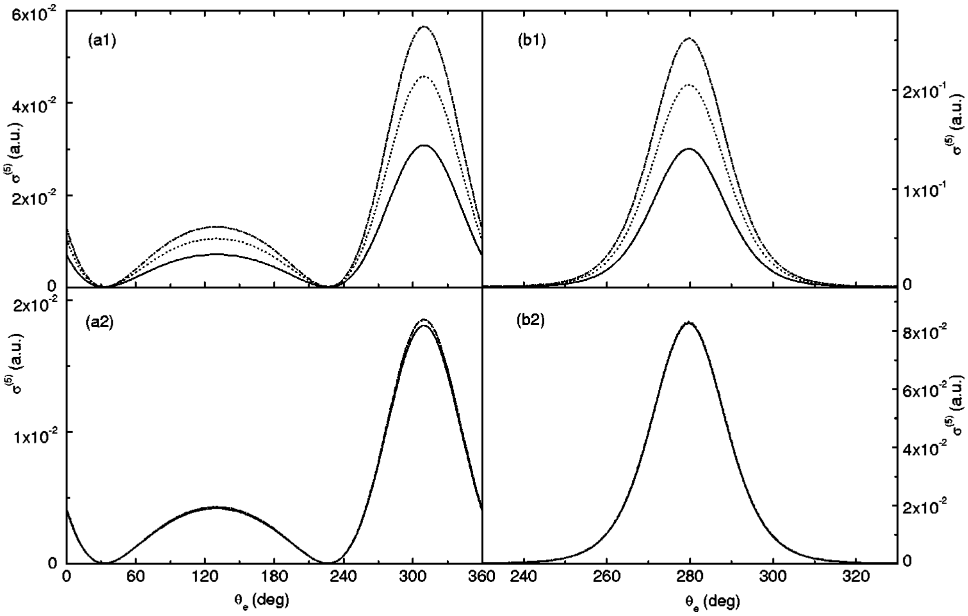

FIG. 1. Fivefold differential cross sections as a function of the ejection angle θ_e for H_2, D_2, and T_2 molecules for an energy resolution of 3 eV. (a1) Results obtained as in *I* for $\theta_s=1°$. (a2) Present results obtained with Eq. (9) for $\theta_s=1°$. (b1) Same as (a1), but $\theta_s=8.9°$. (b2) Same as (a2), but $\theta_s=8.9°$.

(J',M') levels of a given vibrational $(\nu' J')$ level of the ground electronic state of the residual ion:

$$\sigma^{(7)}_{\nu_f,\nu_i,JM} = \sum_{J'M'} 2(2\pi)^4\frac{k_e k_s}{k_i} \times \left|\langle Y_{J'M'}|\int_0^\infty d\rho\, \rho^2 \Xi^*_{\nu_f J'}(\rho)\Theta_{\nu_i J}(\rho) t^e_{if}(\boldsymbol{\rho})|Y_{JM}\rangle\right|^2. \tag{5}$$

As the final rotational levels are not resolved in the existing experimental setups, we can apply the closure relation over the final rotational states by neglecting (following Iijima *et al.* [8] and McCarthy [9]) the J' dependence of the final state vibrational wave function, and obtain

$$\sigma^{(7)}_{\nu_f,\nu_i,JM} = 2(2\pi)^4\frac{k_e k_s}{k_i} \times \langle Y_{JM}|\left|\int_0^\infty d\rho\, \rho^2 \Xi^*_{\nu_f}(\rho)\Theta_{\nu_i J}(\rho) t^e_{if}(\boldsymbol{\rho})\right|^2|Y_{JM}\rangle. \tag{6}$$

Now, as the rotationally averaged cross section is independent of the initial rotational distribution, for a given quantum number J all $|JM\rangle$ states can be considered as equally probable [10]. Therefore, the differential cross section averaged over the initial quantum number M, for a given value of J, can be written as

$$\sigma^{(5)}_{\nu_f,\nu_i J} = \frac{1}{2J+1}\sum_{M=-J}^{J}\sigma^{(5)}_{\nu_f,\nu_i JM}. \tag{7}$$

Keeping in mind that

$$\sum_{M=-J}^{J} Y^*_{JM}(\Omega_\rho) Y_{JM}(\Omega_\rho) = \frac{2J+1}{4\pi}, \tag{8}$$

it follows that

$$\sigma^{(5)}_{\nu_f,\nu_i J} = \frac{1}{4\pi}\int d\Omega_\rho\, 2(2\pi)^4\frac{k_e k_s}{k_i} \times \left|\int_0^\infty d\rho\, \rho^2 \Xi^*_{\nu_f}(\rho)\Theta_{\nu_i J}(\rho) t^e_{if}(\boldsymbol{\rho})\right|^2, \tag{9}$$

which differs, for $J=0$, from the expression used in I obtained by replacing Eq. (1) into Eq. (2). It is interesting to note that the sum over all final rotational states and the average over the initial values of M above performed are equivalent to the point of view adopted in I. In fact, it was previously assumed that $\boldsymbol{\rho}$ remains fixed during the collision process due to the fact that the collision time is much smaller than the vibrational and rotational times. Then, averaging over rotational states is equivalent to integrating over all the orientations of the molecule taken to be equally probable, and then to dividing by 4π.

Initial and final electronic wave functions corresponding to the H_2 molecule and to the $H_2{}^+ + e^-$ system are chosen as in I. According to this, the initial ${}^1\Sigma_g^+$ bound state of the diatomic molecule is given by the Heitler-London wave function, and the final ${}^2\Sigma_g^+$ bound state for the residual molecular ion by a linear combination of atomic orbitals.

In the following we consider $\nu_i=0$ and $J=0$, and for simplicity $\sigma_{\nu_f,00}$ is written as σ_{ν_f}. In Fig. 1, fivefold differential cross sections as a function of the ejection angle θ_e are given for an energy resolution of 3 eV that corresponds to the sum $\sigma^{(5)}_{\nu_f}$ given by Eq. (9) over all final vibrational states. Two different physical situations are considered: the first one is obtained for a small scattering angle $\theta_s=1°$, and the second one corresponds to a larger scattering angle $\theta_s=8.9°$.

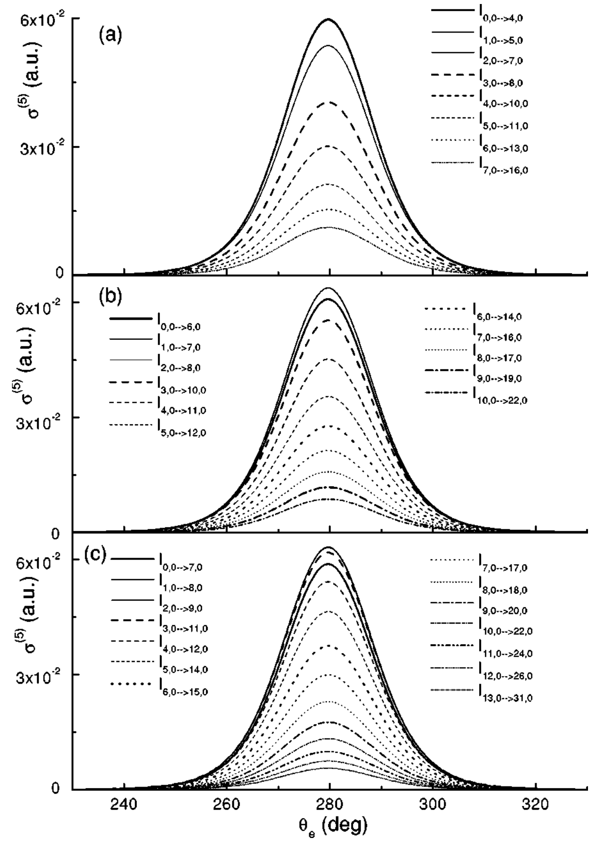

FIG. 2. Fivefold differential cross sections as a function of the ejection angle θ_e for an energy resolution of 1 eV for (a) H_2, (b) D_2, and (c) T_2 molecules. The incidence and ejection energies are 4168 and 100 eV, respectively, and $\theta_s = 8.9°$.

The incident and ejected electron energies are 4168 and 100 eV, respectively. The obtained profiles for H_2, D_2, and T_2 targets are all in close agreement. This behavior is totally different from the one obtained in I by inserting Eq. (1) into Eq. (2) as also shown in the figure.

In Fig. 2, fivefold differential cross sections as a function of θ_e are presented for an energy resolution of 1 eV. The incident and ejection energies are taken as in Fig. 1, and $\theta_s = 8.9°$. For this energy resolution, different bands of final vibrational states must be included depending on the considered molecular target. In this case $\sigma^{(5)}_{\nu_f}$, as defined by Eq. (9), must be summed over the different bands, giving place to the expression

$$\sigma^{(5)}_{(\nu_{f0},\nu_{f0}+\Delta\nu_{f0})} = \sum_{\nu_f=\nu_{f0}}^{\nu_{f0}+\Delta\nu_{f0}} \sigma^{(5)}_{\nu_f}, \tag{10}$$

where ν_{f0} is the lowest final vibrational state of the band, and $\Delta\nu_{f0}$ is the number of considered vibrational states. The lowest bands of the final vibrational states dominate the differential cross sections for H_2, T_2, and D_2 targets. These are the cases for the (0,4) and (1,5) bands in $H_2{}^+$, the (1,7) band in $D_2{}^+$, and the (1,8) and (2,9) bands in $T_2{}^+$. This behavior is again completely different from the one obtained using the crude approximation given by Eq. (1) (see I).

To sum up, $(e,2e)$ multiple differential cross sections have been calculated for a coplanar geometry using the formal scattering theory. The influence of vibrational states on differential cross sections is determined according to the energy resolution. When differential cross sections are summed over all final possible states, the results for different targets are in close agreement. This behavior was obtained for a noncoplanar geometry in Ref. [11], where the theoretical procedure is valid for any kinematics. Moreover, we have estimated separately that for the kinematic conditions considered in this work, the inclusion of all possible final vibrational states in the computation of fivefold differential cross sections gives a contribution of at most 4%, in comparison with a calculation in which only electronic transitions are taken into account.

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