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Electron capture from a hydrogen molecule at a fixed orientation of the molecular axis

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Electron capture in fast-proton–H_2 collisions has been reexamined in the Brinkman-Kramers approximation. The interference, first noted by T. F. Tuan and E. Gerjuoy [Phys. Rev. **117**, 756 (1960)], between two capture amplitudes associated with two centers in the molecule is found to be more pronounced at fixed orientations of the molecular axis as compared to the results obtained by averaging over all orientations. This interference effect varies significantly with the orientations of the molecule. For example, in a certain angular range the number of oscillations in the differential cross sections (DCS) is maximum at $\theta_\rho=90°$, $\phi_\rho=0°$, where θ_ρ and ϕ_ρ are the polar and azimuthal angles, respectively, of the molecular axis with respect to the incident-beam direction. The number of oscillations of the DCS decreases (i) as impact energy decreases at a fixed value of orientation and (ii) as the orientation changes from the perpendicular position ($\theta_\rho=90°$, $\phi_\rho=0°$) to other orientations for a fixed proton energy. This interference effect has not yet been observed experimentally.

Electron capture from molecular targets differs from capture from single atoms because of the multicenter configuration of molecules. For example, at high impact energies, a diatomic molecule may be regarded as a target where the incoming-projectile wave fronts scatter from both atomic centers producing interference[1] patterns in the outgoing waves. This idea was recognized by Tuan and Gerjuoy[2] who first calculated electron-capture cross sections from molecular hydrogen by protons at high collision velocities. However, Tuan and Gerjuoy[2] and later Band,[3] and Ray and Saha[4,5] averaged the cross sections over all the orientations of the molecule. As a consequence, the interference effects were not readily apparent. In this paper we evaluate differential cross sections as a function of fixed molecular orientations and find an interference structure similar to the "classical" Young's two-slit interference pattern. Experimental observation using a Coulomb explosion technique appears to be feasible.[6] For $p+H_2$, we illustrate how this interference pattern varies with the orientation of the target molecular axis and with the velocity of the projectile.

Compared to the number of calculations done for electron capture from atoms,[7] the number of calculations for capture from molecules[2–5,8–10] is relatively small. At high collision velocities, which we consider, Tuan and Gerjuoy first formulated the theory and evaluated cross sections for the prototype $p+H_2$ system using the first-order approximation[11,12] credited to Brinkman and Kramers (BK). Tuan and Gerjuoy found that it is not generally correct to consider H_2 as two H atoms. Later Ray and Saha[4] repeated the Tuan and Gerjuoy (in the BK approximation) calculations and obtained quantitatively different results from Tuan and Gerjuoy, although the overall conclusions of Tuan and Gerjuoy remained intact.

We use notation of Tuan and Gerjuoy[2] to describe the orientation dependence of the cross sections for the charge transfer process,

$$p+H_2 \rightarrow H+H_2{}^+ \ . \tag{1}$$

The full transition matrix element for electron capture by protons from hydrogen molecule is given by[2]

$$\begin{aligned}\langle \psi_b \mid V_i \mid \psi_a \rangle = {} & \exp(-i\boldsymbol{\alpha}\cdot\boldsymbol{\rho}/2) \\ & \times \int \exp(-i\boldsymbol{\alpha}\cdot\mathbf{r}_1)\exp(i\boldsymbol{\beta}\cdot\mathbf{r}'_1)\phi_i^*(\mathbf{r}_2;\boldsymbol{\rho}) \\ & \times u^*(\mathbf{r}'_1)V_i\phi_m(\mathbf{r}_1,\mathbf{r}_2;\boldsymbol{\rho})d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}'_1 \ ,\end{aligned} \tag{2a}$$

where

$$\begin{aligned}&\boldsymbol{\alpha}=\mathbf{K}_b-\mathbf{K}_a, \quad \boldsymbol{\beta}=[M/(m+M)]\mathbf{K}_b-\mathbf{K}_a \ , \\ &V_i=V_{1C}+V_{AC}+V_{BC}+V_{2C} \ ,\end{aligned} \tag{2b}$$

with $\mathbf{K}_a$ and $\mathbf{K}_b$ initial and final momenta, $\mathbf{r}_1$ and $\mathbf{r}_2$ the coordinates of electrons 1 and 2 with respect to the centers A and B, respectively; $\mathbf{r}'_1$ is the coordinate of electron 1 with respect to the projectile C and $\boldsymbol{\rho}$ is the internuclear vector of the molecule. The ϕ_m and ϕ_i are, respectively, the molecular and ionic wave functions. In general, three kinds of molecular wave functions may be used: the extreme atomic orbitals of Wang,[13] the extreme molecular orbital or the intermediate Weinbaum[14] wave functions. The general forms of the molecular and ionic wave functions shown in Eq. (2a) are given by

$$\begin{aligned}\phi_m(\mathbf{r}_1,\mathbf{r}_2,\boldsymbol{\rho})=N_m\{ & u_{mA}(1)u_{mB}(2)+u_{mA}(2)u_{mB}(1) \\ & +C[u_{mA}(1)u_{mA}(2) \\ & +u_{mB}(1)u_{mB}(2)]\} \ , \\ \phi_i(\mathbf{r}_2;\boldsymbol{\rho})=N_i^{\pm}[& u_{iA}(2)\pm u_{iB}(2)] \ ,\end{aligned} \tag{3a}$$

where

$$\begin{aligned}&N_m=1/\{2[(1+c^2)(1+\Delta_m^2)+4c\Delta_m]\}^{1/2} \ , \\ &N_i^{\pm}=1/[2(1\pm\Delta_i)]^{1/2} \ ,\end{aligned}$$

and (3b)

$$\Delta_m = (1 + Z_m\rho + \tfrac{1}{3}Z_m^2\rho^2)\exp(-Z_m\rho) ,$$

$$\Delta_i = (1 + Z_i\rho + \tfrac{1}{3}Z_i^2\rho^2)\exp(-Z_i\rho) .$$

In Eqs. (2a) and (3a), u_m, u_i, and u are the ground-state hydrogenic wave functions with effective charges Z_m, Z_i, and 1, respectively. Values of c, Z_m, and Z_i are different in different types of target wave functions ϕ_m and ϕ_i. In our calculation, we use the intermediate Weinbaum[14] wave functions with values $c = 0.256$, $Z_m = 1.193$, and $Z_i = 1.4$.

The molecular analog (MBK) of the atomic amplitude for the 1s-1s capture is then taken to be

$$\langle \psi_b \mid V_i \mid \psi_a \rangle \mid_{\mathrm{MBK}} = I_{\mathrm{BK}}(A) + I_{\mathrm{BK}}(B) , \tag{4a}$$

where

$$I_{\mathrm{BK}}(A) + I_{\mathrm{BK}}(B) = (e^{i\alpha\cdot\rho/2} \pm e^{-i\alpha\cdot\rho/2})N_i^{\pm}N_m \times (1 \pm c)\Delta_{im} + (c \pm 1)\chi_{im}]I_{pe} , \tag{4b}$$

with

$$\Delta_{im} = 8(Z_iZ_m)^{3/2}/(Z_i + Z_m)^3 ,$$

$$\chi_{im} = \frac{8(Z_iZ_m)^{3/2}}{\rho\nu^3}[Z_i(\rho\nu - 4Z_m)e^{-Z_m\rho} + Z_m(\rho\nu + 4Z_i)e^{-Z_i\rho}] ,$$

$$\nu = Z_i^2 - Z_m^2 , \tag{4c}$$

$$I_{pe} = \frac{32\pi Z_m^{5/2}}{(\beta^2 + 1)(\alpha^2 + Z_m^2)^2} .$$

In Eq. (4b) it is the term $e^{i\alpha\cdot\rho/2} \pm e^{-i\alpha\cdot\rho/2}$, which is responsible for the orientation dependence (i.e., θ_ρ, ϕ_ρ) of the capture cross sections. The differential cross section at a fixed orientation is given by

$$\frac{d\sigma}{d\Omega_s d\Omega_\rho} = \frac{2\mu_a\mu_b K_b}{(2\pi)^2 K_a} \mid I_{\mathrm{BK}}(A) + I_{\mathrm{BK}}(B) \mid^2 . \tag{5}$$

Substituting Eq. (4b) in Eq. (5) we obtain

$$\frac{d\sigma}{d\Omega_s d\Omega_\rho} = \frac{4\mu_a\mu_b K_b}{(2\pi)^2 K_a}(N_i^{\pm}N_m)^2[(1 \pm c)\Delta_{im} + (c \pm 1)\chi_{im}]^2 \times I_{pe}^2[1 \pm \cos(\alpha\cdot\rho)] . \tag{6}$$

Here

$$\alpha\cdot\rho = K_b\rho[\cos\theta_s\cos\theta_\rho + \sin\theta_s\sin\theta_\rho\cos(\phi_s - \phi_\rho)] - K_a\rho\cos\theta_\rho , \tag{7}$$

with (θ_s, ϕ_s) being the scattering polar and azimuthal angles. If ϕ_s is not zero, then the system is not cylindrically symmetric; however, we can still assume ϕ_s to be zero with respect to beam axis in the laboratory frame. It is also to be noted that I_{pe} is a smoothly varying function of θ_s. In the above expressions the ± signs correspond, respectively, to the gerade and ungerade states of the H_2^+ ion. The interference effects which we emphasize in this paper are due to the $[1 \pm \cos(\alpha\cdot\rho)]$ term[1] in Eq. (6).

We first consider differential cross sections for electron capture from H_2 by high-velocity protons as a function of orientation (θ_ρ, ϕ_ρ) of the molecular axis with respect to beam axis. A typical result is shown in Fig. 1 where we compare capture from H_2 at a fixed orientation of (90°,0°). The azimuthal angles of the molecule and scattered proton, ϕ_ρ and ϕ_s, are both taken to be zero. In Fig. 1 we show capture cross sections only from the gerade state of H_2 (capture from the ungerade state is relatively small). For a particular orientation of the molecule, oscillations from the $1 + \cos(\alpha\cdot\rho)$ factor in Eq. (6) are evident in Fig. 1. If one sums over all orientations of the H_2 molecule the oscillations disappear. As mentioned earlier, this sum over all orientations was done by all previous authors.[2–5] We confirm all the results reported by Ray and Saha[4,5] for the differential as well as total cross sections. We also include in Fig. 1 the capture cross sections from two H atoms corresponding to the independent electron approximation. The interference structure represents a deviation from the independent electron approximation. The shape of the differential cross sections for capture from atomic hydrogen is similar to the shape of the envelope of the oscillations predicted for capture from H_2.

In Fig. 2 we present differential cross sections at different orientations of the H_2 molecule at a fixed collision energy of 500 keV. The three cases shown are for $\theta_\rho = 90°$, 45°, and 0° with $\phi_s = \phi_\rho = 0$. As the angle θ_ρ decreases from the perpendicular direction (i.e., $\theta_\rho = 90°$) to smaller angles, the pattern of oscillations changes: the spacing between two successive dips widens, thus reducing the number of oscillations in a fixed angular range. For example, at $\theta_\rho = 20°$ and 5° values the number of dips in this angular range is 2 and 1, respectively. For ρ perpendicular to the beam direction, we have $\alpha\cdot\rho = \rho K_b \sin\theta_s$

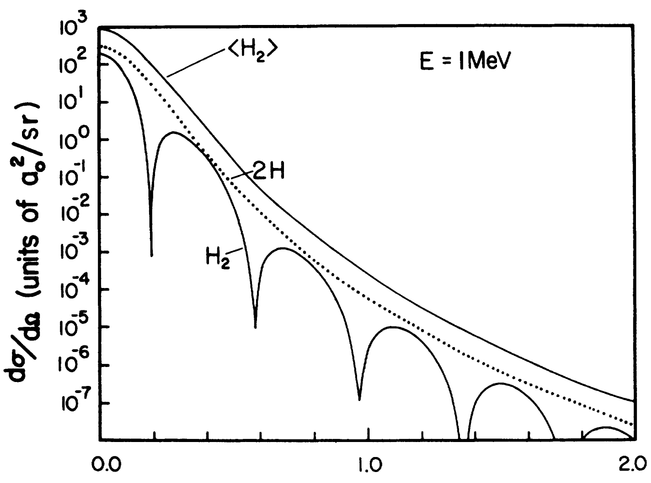

FIG. 1. Differential cross sections (in units of a_0^2/sr) at a fixed energy 1 MeV: $\langle H_2 \rangle$, $p + H_2$ cross section averaged over all possible orientation; H_2, $p + H_2$ cross section for a fixed orientation $\theta_\rho = 90°$, $\phi_\rho = 0°$; 2H, the cross sections for $p + H$ multiplied by two.

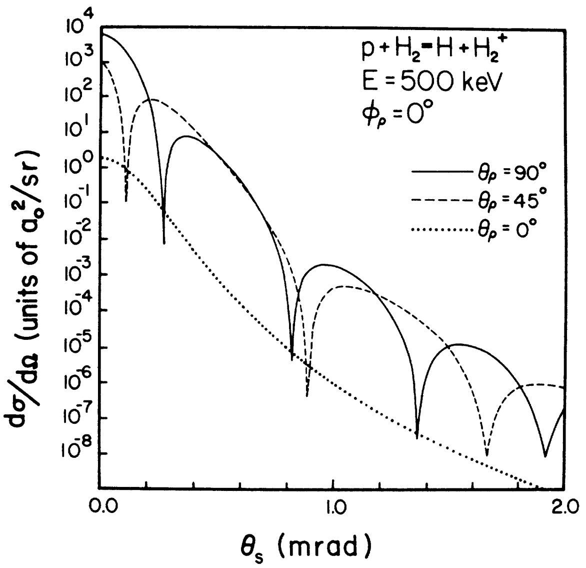

FIG. 2. Differential cross sections (in units of a_0^2/sr) for a fixed projectile energy 500 keV at different orientations of the molecular axis: ——, $\theta_p=90°$, $\phi_p=0°$; — — —, $\theta_p=45°$, $\phi_p=0°$; · · · ·, $\theta_p=0°$, $\phi_p=0°$.

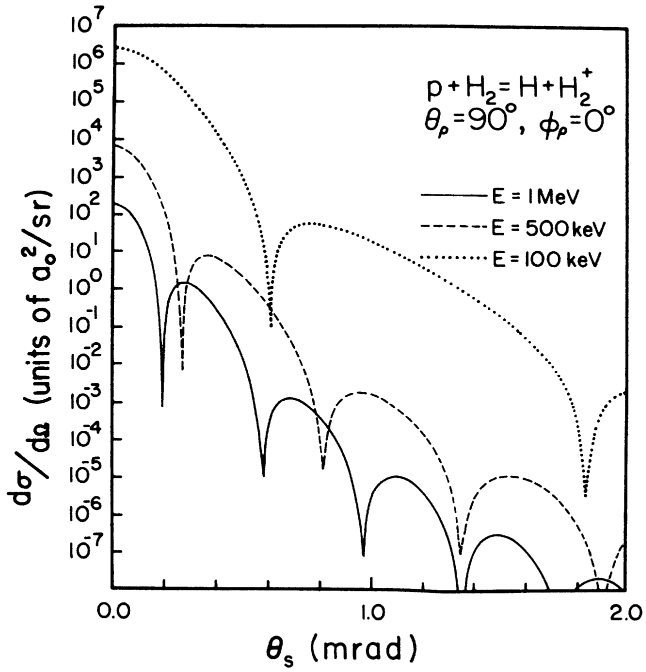

FIG. 3. Differential cross sections (in units of a_0^3/sr) for a fixed orientation $\theta_p=90°$, $\phi_p=0°$ of the molecular axis at various energies: ——, 1 MeV; — — —, 500 keV; · · · ·, 100 keV.

which varies linearly in θ_s for forward angles. For $\theta_p=0°$ or 180°, i.e., when $\boldsymbol{\rho}$ is parallel to the beam direction, $\boldsymbol{\alpha}\cdot\boldsymbol{\rho}=\pm(K_b\rho\cos\theta_s-K_a\rho)$, which is independent of θ_s for small θ_s since $K_a\neq K_b$. We note that at an orientation where $\boldsymbol{\alpha}\cdot\boldsymbol{\rho}=\pi$ a minimum will occur at a scattering angle given by the relation,

$$\cos\theta_s=(\pm\pi+K_a\rho)/K_b\rho \ ,$$

where the plus and minus signs correspond to $\theta_p=0°$ and $\theta_p=180°$, respectively. At these orientations the cross section integrated over scattering angles will be smaller than at other orientations.

We now explore the energy dependence of the differential cross sections at a fixed orientation. We plot the differential cross sections for a fixed molecular orientation of $\theta_p=90°$ and $\phi_p=0°$ at various projectile energies in Fig. 3. As expected, we see from Fig. 3 that as the projectile energy increases the differential cross sections decrease rapidly except near the minima positions. The number of oscillations in a given angular range increases with the increasing collision energy since the wavelength of the incoming wave is decreasing.

In our calculation the interference phenomenon in the differential cross section curves is analogous[1] to the "classical" two-slit scattering. If the H_2 molecules were replaced by an evenly spaced array of atoms, then the interference maxima would become sharper as the size of the array was increased. In one dimension this corresponds to scattering of waves by a diffraction grating. For an infinite two-dimensional array, the interference pattern would be similar to that of neutron diffraction by crystals. In such a system one expects that capture into states of different final energy, hence different momentum transfer, to scatter primarily into distinct angles. In principle, atoms with electrons in separate well-defined states could be produced in well-defined directions.

The present BK calculation is the simplest possible quantum calculation for this system. For capture from atoms the BK differential cross sections are in good agreement with the shape of the observed angular dependence at forward angles. However, the normalization, i.e., the total BK cross section, is typically at least a factor of 3 too large. Improved calculations of capture from molecular hydrogen are possible. This includes better first-order approximations,[15–17] higher-order approximations,[15–21] and the coupled-channel method.[7,22] The higher-order methods tend to include phase terms not included in our simple first-order BK method. Such higher-order phase corrections could modify the interference pattern quantitatively but the general behavior of the oscillations and the "classical" analogy with the two-slit experiment should not change since the interference term $1+\cos(\boldsymbol{\alpha}\cdot\boldsymbol{\rho})$ does not depend upon the scattering amplitude and the BK approximation has been successful at predicting shapes of differential cross sections at forward angles. Of course, at energies below 25 keV, where molecular orbital effects including alignment and transfer to excited states could be significant, these high-energy predictions may not be accurate. We note that we do not expect small fluctuations in the monochromaticity of the beam to alter the interference significantly. And we do not expect that exact symmetry of the two centers is required for interference to occur.

An experimental observation of the differential cross sections at a fixed molecular orientation would directly test the interference phenomenon discussed in this paper. It has been suggested[6] that the molecular orientation may be selected by observing recoiling molecular fragments. Although such an experiment may be difficult to perform, it would put to the test our rather simple and appealing picture where scattering from a molecule is regarded as wave scattering from two localized atomic centers.

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