RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 43, NUMBER 5 1 MARCH 1991

Rapid Communications

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Electric dipole moments of $H(n=2)$ induced in H^+-He and H-He collisions

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(Received 13 August 1990)

Electric dipole moments of $H(n=2)$ induced by H^+-He and H-He collisions are investigated at incident energies of 5–25 keV. For these collision systems, an energy-dependent forward-backward asymmetry (dipole moment) of the emitted Lyman-α radiation as a function of an external electric field is observed. In H^+-He collisions, the measured dipole moment is positive, corresponding to an electron trailing behind the proton. The same analysis applied to the H-He system shows the electron riding in front of the proton. The observed differences between H and H^+ impact are partly explained with the help of a simple model which assumes different cross sections for excitation into the different Stark states.

The excitation of a hydrogen atom to the $H(n=2)$ states is a prototype case for the study of coherent excitation mechanisms operating during atomic collision processes. This is because of the near degeneracy among the different $H(n=2)$ levels, for which, due to the small energy splitting $\Delta E \lesssim 10^{-5}$ eV, significant differences in the time development occur after $\tau = \hbar/\Delta E \approx 10^{-11}$ s ($\hbar$ is Planck's constant); this time is considerably larger than typical collision times of $\tau_{\rm col} \approx 10^{-16}$ s. In this Rapid Communication we present and discuss new results for the electric dipole moment induced in the excited $H(n=2)$ state by (positively charged) hydrogen ion impact, $H^+ + He \rightarrow H(n=2) + He^+$, and by (neutral) hydrogen atom impact, $H + He \rightarrow H(n=2) + He$. Since in the present experiment we integrate over all scattering angles of the projectile, the excited $H(n=2)$ state is best described in terms of a density matrix[1] $\rho_{n=2}$ at the instant of the collision ($t=0$):

$$\rho_{n=2} = \begin{bmatrix} \sigma_{2s_0} & 0 & \sigma_{sp} & 0 \\ 0 & \sigma_{2p_{-1}} & 0 & 0 \\ \sigma_{sp}^* & 0 & \sigma_{2p_0} & 0 \\ 0 & 0 & 0 & \sigma_{2p_{+1}} \end{bmatrix}, \quad (1)$$

where the $\sigma_{lm} = \langle |f_{lm}|^2 \rangle$ are the partial cross sections for excitation into the $H(2s)$ and $H(2p_m)$ ($m=0,\pm 1$) substates, f_{lm} is the corresponding excitation amplitude, $\sigma_{sp} = \langle f_{00} f_{10}^* \rangle$ is the so-called *s-p* coherence, and where $\langle \cdots \rangle$ denotes an integration over all scattering angles. An electric dipole moment corresponds to a shift of the center of charge with respect to the center of mass (for example, an electron trailing behind a proton) and thus manifests itself in a forward-backward asymmetry with regard to an external electric field;[2] it is related to the real part $\mathrm{Re}(\sigma_{sp})$ of the *s-p* coherence.[3] Collision-induced dipole moments $\langle D \rangle_z$ have, for example, been observed previously for charge-exchange excitation of $H(n=2)$ and $H(n=3)$ in H^+-He collisions at incident energies above 20 keV.[4,5] The imaginary part $\mathrm{Im}(\sigma_{sp})$ is related to the product $\langle \mathbf{L} \times \mathbf{A} \rangle_z$, where $\mathbf{L}$ and $\mathbf{A}$ are the angular momentum vector and the Runge-Lenz vector, respectively; classically, the vector $\mathbf{L} \times \mathbf{A}$ points along the orbital electron velocity at the perihelion of its Kepler orbit.[3,4] Hence, we have

$$\langle D \rangle_z = \frac{6\,\mathrm{Re}(\sigma_{sp})}{\sigma_t}, \quad \langle \mathbf{L} \times \mathbf{A} \rangle_z = -\frac{2\,\mathrm{Im}(\sigma_{sp})}{\sigma_t}, \quad (2)$$

where $\sigma_t = \sum \sigma_{lm}$ is the total cross section for $H(n=2)$ excitation. To extract all the relevant elements of the $\rho_{n=2}$ density matrix, we utilize Stark mixing of the $H(2s)$ and $H(2p)$ states in an external electric field and measure (perpendicular to the direction of the electric field and as a function of the field strength) the linear and circular polarizations of Lyman-α radiation ($\lambda = 121.6$ nm) emitted during the decay of the excited $H(n=2)$ state to the $H(1s)$ ground state. The linear polarization $P_l = (I_\parallel - I_\perp)/(I_\parallel + I_\perp)$ was investigated for axial (with respect to the projectile direction, z axis) electric fields, with $I_\parallel$ and $I_\perp$ the integrated light intensities with the electric vector parallel and perpendicular, respectively, to the z axis. These measurements were performed with the help of a Brewster-type linear polarizer; i.e., we used a lithium-fluoride (LiF) plate inclined at $\sim 60°$ with respect to the optical axis resulting in a polarization sensitivity of 90%. The circular polarization $P_c = (I_- - I_+)/(I_- + I_+)$, where I_+ and I_- are the integrated light intensities with positive and negative helicity, respectively, was measured for transverse electric fields. For circular polarization measurements, a quarter wave plate made from magnesium fluoride (MgF_2) was inserted in front of the linear polarizer; its polarization sensitivity is ~98%. Details of the apparatus have been described previously[6,7] and shall not be repeated here.

The measured degree of linear polarization P_l for 20-keV H^+-He collisions is displayed in Fig. 1; it shows a pronounced forward-backward asymmetry which is due to the collision-induced electric dipole moment. In the following, we concentrate on the evaluation of σ_{sp}; details of the evaluation procedure and results for the partial substate cross sections will be published in a forthcoming publication.[8] To extract the real part $\mathrm{Re}(\sigma_{sp})$ from the measured P_l, we (i) have to expand the initial state Ψ in terms of Stark states $|k\rangle$, (ii) take the time evolution of the Stark states into account, and (iii) calculate matrix elements for the radiative decay of these states to the H(1s) ground state. The corresponding intensities $I_\parallel(t)$ and $I_\perp(t)$ of the emitted light with electric vector parallel and perpendicular, respectively, to the z axis are calculated using

$$I_{\mathbf{r}}(t)=\left|\sum_{k=1}^{4}\langle 1s_{1/2}|e\mathbf{r}|k\rangle e^{(i\omega_k-\gamma_k/2)t}\langle k|\Psi\rangle\right|^2, \quad (3)$$

where $\hbar\omega_k$ and γ_k are the electronic binding energy and the radiative width, respectively, of the $|k\rangle$ Stark state. Following Lüders,[9] the (relatively weak) external field causes a mixing of the unperturbed hydrogenic states $\Phi_i=|jm_j\rangle$; the perturbed $H(n=2)$ eigenstates (Stark states) $|k\rangle$ are then given as linear combinations $|k\rangle=\sum a_{ki}\Phi_i$, where the a_{ki} ($k=1,\ldots,4$) are expansion coefficients which depend on the electric-field strength F.[10] The time-dependent intensities $I(t)$ have been integrated over the finite observation interval over which production and decay of the excited $H(n=2)$ atoms occurs to yield the integrated intensities $I_\parallel$ and $I_\perp$; this smears out the rapid intensity oscillations (Stark beats) which were not observable in the present experiment.[11,12]

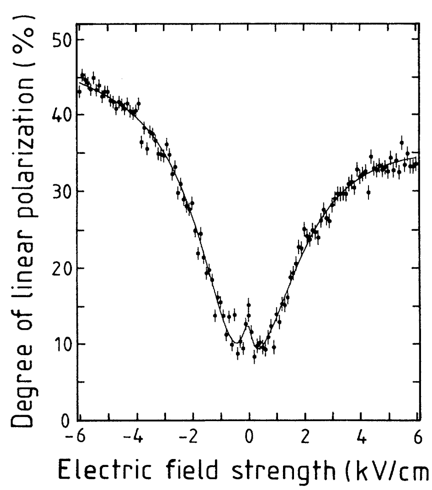

FIG. 1. Degree of linear polarization vs electric-field strength for 20-keV H^+-He collision; the solid line is a least-squares fit to the data (see text).

The measured linear polarization was fitted according to Eq. (3) and the dipole moment $\langle D\rangle_z$ was extracted. In Fig. 2 we display our results; in general, $\langle D\rangle_z$ is small around incident energies of 5–15 keV and rises towards higher energies. The sign of the measured dipole moment indicates that the captured electron is lagging behind the proton. This result is intuitively acceptable, as it corresponds to an electron which is attracted by both the incident proton and by the product helium ion and, therefore, tends to stay in-between. This argument seems to be further supported by measurements for the neutral H-He collision system, where significantly smaller dipole moments and of opposite sign are observed (Fig. 2). In order to illustrate the origin of the collision-induced dipole moment $\langle D\rangle_z$ on a more quantitative basis, we express the coherently excited $H(n=2)$ wave function Ψ at $t=0$ as

$$\Psi=f_+|+\rangle+f_-|-\rangle+f_1|2p_1\rangle+f_{-1}|2p_{-1}\rangle \quad (4)$$

where $|\pm\rangle=\mp(1/\sqrt{2})(|2s_0\rangle\pm|2p_0\rangle)$ are the (large-field) Stark states, and where $f_\pm=\mp(1/\sqrt{2})(f_{00}\pm f_{10})$ are the corresponding excitation amplitudes. Rewriting Eq. (2) we obtain

$$\langle D\rangle_z=\frac{3(\sigma_+-\sigma_-)}{\sigma_t}, \quad (5a)$$

$$\langle \mathbf{L}\times\mathbf{A}\rangle_z=-\frac{2\sqrt{\sigma_+\sigma_-}}{\sigma_t}\sin\bar{\chi}, \quad (5b)$$

with $\bar{\chi}$ the average phase between f_+ and f_-. The two Stark states $|\pm\rangle$ possess an electric dipole moment $d=\pm 3ea_0$ (e is the electron charge, and a_0 is the Bohr radius); a net dipole moment hence results, if the two excitation cross sections $\sigma_\pm=\langle|f_\pm|^2\rangle$ are not equal. Unequal cross sections may result, for example, if the two excitation energies E_+ and E_- are different. Since during the collision the atomic states are considerably perturbed

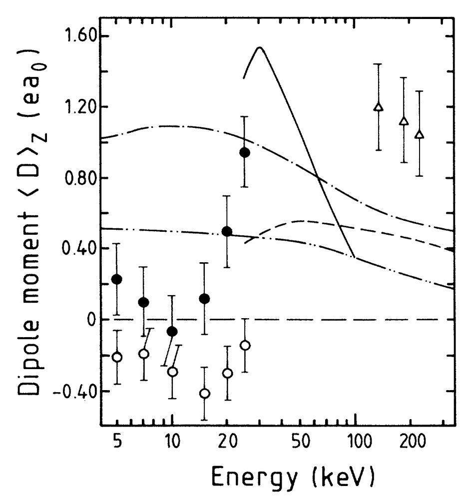

FIG. 2. Dipole moment $\langle D\rangle_z$ for $H(n=2)$ excitation in H^+-He collisions: ●, present results; △, Ref. 4; AO+ calculations (solid line, Ref. 3); CDW-PCI calculations (dashed line, Ref. 4); present model (dash-dotted line); H-He collisions: ○, present results; present model (dash-two-dotted line) (see text).

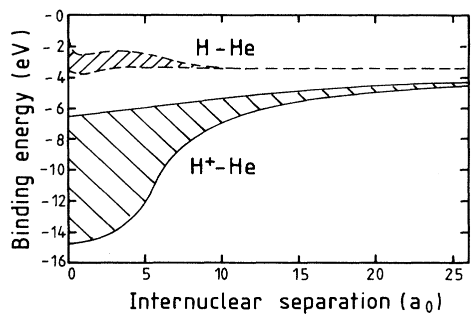

FIG. 3. Binding energies of the relevant quasimolecular states correlating with the $|+\rangle$ and $|-\rangle$ Stark states in H$^+$-He (solid lines) and H-He (dashed lines) collisions estimated from calculated molecular energies. (Refs. 14 and 17).

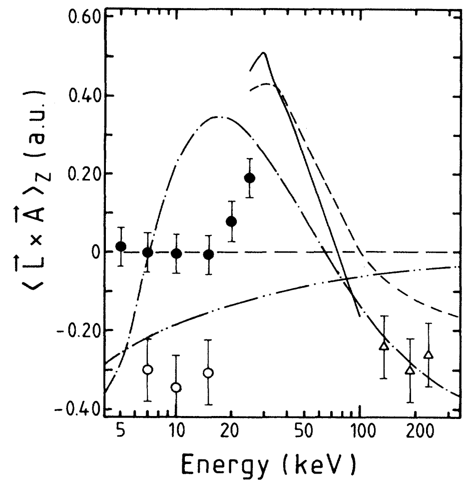

FIG. 4. The moment $\langle \mathbf{L}\times\mathbf{A}\rangle_z$ for H($n=2$) excitation in H$^+$-He collisions: ●, present results; △, Ref. 4; AO+ calculations (solid line, Ref. 3); CDW-PCI calculations (dashed line, Ref. 4); present model (dash-dotted line); H-He collisions: ○, present results; present model (dash-two-dotted line) (see text).

by the presence of the other collision partner, significant energy splittings among formerly degenerate electronic states may arise. At low incident energies, the two collision partners form a transient quasimolecule;[13] for the singlet (H-He)$^+$ system we use calculated molecular energies[14] to estimate the energy splitting among the diabatic $2s\sigma$ and $3p\sigma$ molecular states[15] which, for large internuclear separations $R\rightarrow\infty$, correlate with the $|+\rangle$ and $|-\rangle$ Stark states, respectively (Fig. 3). Qualitatively, due to the largely different excitation energies in the (H-He)$^+$ system, we expect significantly different cross sections σ_+ and σ_-. As estimates of σ_+ and σ_- we use the Oppenheimer-Brinkman-Kramers (OBK) approximation,[16] using at low incident velocities the molecular $2s\sigma$ and $3p\sigma$ energies at $R=0$. At large incident velocities, due to the short interaction time, the atomic states experience little perturbation and we have to take the energies of the unperturbed states. In between, we average the molecular energies over the typical response time $\tau_s=\hbar/E_s$ of the atomic system to obtain average energies

$$\bar{E}_s=\frac{1}{\tau_s}\int_{-\tau_s/2}^{\tau_s/2}E_s[R(t)]dt\ .$$

The estimated dipole moment $\langle D\rangle_z$ is displayed in Fig. 2 and shows the expected behavior: it is large at small incident velocities and approaches zero for high velocities. Qualitatively, the predicted behavior is in reasonable agreement with predictions from the continuum-distorted-wave approximation accounting for post-collision-interaction effects (CDW-PCI, Ref. 4) and with the atomic-orbital expansion (AO+) calculations of Jain, Lin, and Fritsch.[3] The same analysis applied to the H-He system[17] shows that the relevant energy splitting is considerably smaller; the predicted $\langle D\rangle_z$ is, hence, less pronounced although still significantly larger than zero (Fig. 2).[18] Our simple model qualitatively explains the observed differences between (H-He)$^+$ and H-He collisions. Its apparent failure below $\lesssim 20$ keV is most likely due to the radial couplings[14,17] which operate in the transient quasimolecule but have not been considered here.

In Fig. 4 we display our results for $\langle \mathbf{L}\times\mathbf{A}\rangle_z$ which were extracted from measurements of the circular polarization P_c in transverse fields. As before the dipole moment, $\langle \mathbf{L}\times\mathbf{A}\rangle_z$ is small in H$^+$-He collisions up until 15 keV and rises towards larger energies. In connection with previous measurements of DeSerio *et al.*,[4] our data indicate an oscillatory behavior whose origin becomes evident if we realize that the relative phase $\bar{\chi}$ is at least in part due to the accumulated phase difference

$$\Delta\phi=\frac{1}{\hbar v}\int_{R_0}^{\infty}\Delta E(R)dR$$

among the $|+\rangle$ and $|-\rangle$ substates. This quantity is thus sensitive to the very details of the collision process. The fair agreement with the present model is fortuitous, however, since a constant phase $\phi_0=\pi/2$ which possibly arises from the above-mentioned radial couplings was added to the calculated $\Delta\phi$. In H-He collisions, $\langle \mathbf{L}\times\mathbf{A}\rangle_z$, as expected because of the considerably smaller $\Delta\phi$, is negative at the energies considered here and in fair agreement with our calculation.

Note added in proof. Recent calculations by Shingal and Lin[19] confirm the general trends of the present experimental results for H$^+$-He.

This work was supported by the Deutsche Forschungsgemeinschaft in Sonderforschungsbereich 216.

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