Alignment in Molecular Excitation for 3.22-keV H_2^+-He Collisions

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Electronic alignment in molecular excitation, relative to a specific internuclear axis orientation, has been observed for the first time, enabling specific detailed checks of the quasidiatomic molecular-orbital model in kiloelectronvolt atom-molecule collisions. The alignment was determined from the polarization of L_α radiation, resulting from the collision process $H_2^+ + He \rightarrow H_2^{+*} + He \rightarrow H^+ + H(2p) + He$, for specific laboratory scattering angles of H^+. The results suggest that, as in many ion-atom systems, rotational coupling is the dominant excitation mechanism.

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Since the early experimental work of Ziemba *et al.*,[1] and the subsequent theoretical framework of Fano and Lichten,[2] great strides have been made in our understanding of a range of atomic collision processes. Recent work of Dowek *et al.*[3] has successfully extended some of the basic ideas that have proven so valuable in describing binary systems to triatomic systems, that is, to ion/atom-molecule collisions. Specifically, they have been able to provide insights into the inelastic processes of He- and He^+-$H_2(D_2)$ collisions, by incorporating a quasidiatomic molecular-orbital (MO) correlation diagram to describe the collision, although the system is a triatomic one. In addition, they have presented three-dimensional correlation surfaces for the He-H_2 system. The quasidiatomic MO correlation diagram used by Dowek *et al.* is shown in Fig. 1. In this figure the H_2-He MO diagram is constructed in analogy to H-He; one expects that this should give a reasonable picture of the possible electron promotion mechanisms, when the H_2^+ internuclear separation is smaller than the distance between He and H_2^+ centers of mass. Clearly, our understanding of inelastic processes will be greatly advanced if the limits of this generalization of the diatomic to the triatomic can be established.

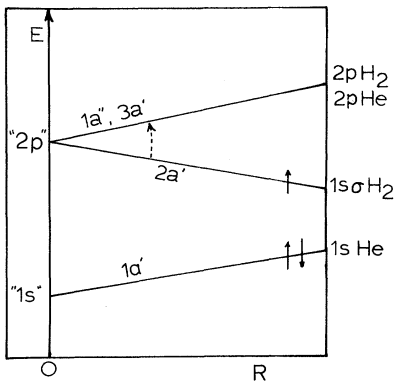

FIG. 1. Quasidiatomic MO diagram for the He-H_2 system. The two arrows on curve $1a'$ refer to the two electrons of He and the one on curve $2a'$ shows the only electron of H_2^+. The $2p$ He and $2p$ H_2 states, being nearly degenerate, are not shown separately. The experimental results suggest that $1s\sigma$ H_2^+ (curve $2a'$) is promoted to $2p\pi$ H_2^+ (curve $3a'$) by rotational coupling.

We wish to report new measurements that indicate the general efficacy of this MO correlation picture and point out how it can be used to understand a range of inelastic processes in $H_2^+ + He$ collisions.

The specific experiment that we have performed is the measurement of the polarization of L_α radiation in coincidence with the scattered proton that results from the process $H_2^+ + He \rightarrow H_2^{+*} + He \rightarrow H^+ + H(2p) + He$. For distant collisions that result in negligible deflection of the H_2^+ c.m., the direction of the H^+ provides information about the final orientation of the H_2^+ internuclear axis and the c.m. H^+ velocity, while the L_α polarization provides information about the alignment of $H(2p)$ relative to that axis.

Details of the apparatus have been discussed elsewhere.[4] The essential features of the apparatus are a needle gas jet to provide a target, a solar-blind L_α detector, and a LiF Brewster-angle polarizer. The L_α photons are detected in a direction perpendicular to a plane defined by the momenta of the incident H_2^+ beam and the scattered H^+ as indicated in Fig. 2.

The measured L_α intensities $I(\beta)$, for specific polarization angles β at specific H^+ scattering angles, as determined by coincidence measurements, are shown in Fig. 3 along with the least-squares fit to the data of a general dipole intensity pattern. The measurements indicate that as the scattering angle is increased, the polarization distribution becomes increasingly "pinched" until at $\theta = \pm 3.25$, the ratio of I_{max}/I_{min} is the maximum allowed by the atomic precession due

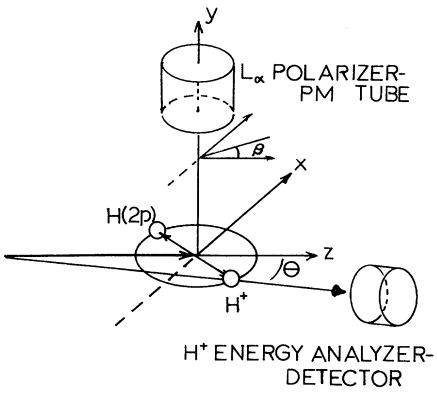

FIG. 2. Schematic arrangement for $H^+ - L_\alpha$ coincidence measurement.

to spin-orbit coupling.[5]

Also schematically shown in Fig. 3 is the range of H_2^+ internuclear-axis orientations at angles α relative to the beam direction that contribute to the H^+-L_α coincidence signal. The energy analyzer does not have sufficient resolution to distinguish different H^+ laboratory velocity vectors

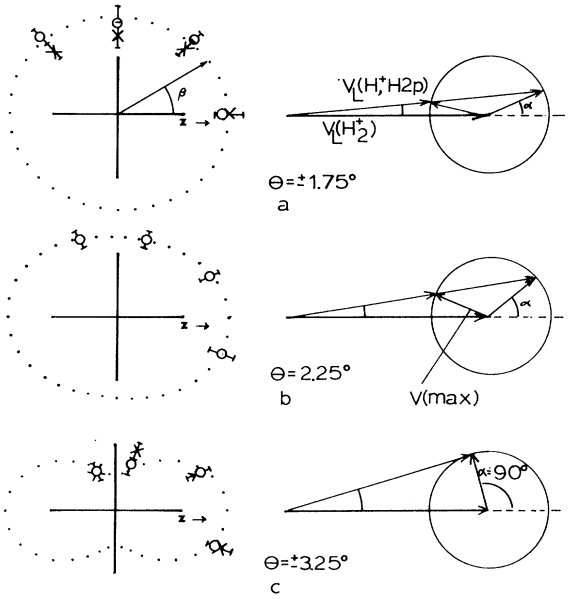

FIG. 3. Left-hand side: Polarization measurements $I(\beta)$ at different H^+ scattering angles for 3.22-keV H_2^+-He collisions. The beam direction is shown by z. Right-hand side: Velocity diagrams showing the variation in range of H_2^+ axis orientations which give rise to the observed polarization patterns as a function of H^+ laboratory scattering angle. The speed of the dissociation products being much smaller than that of the center of mass, the radii of the circles are greatly exaggerated and $\alpha = 90°$ is schematically shown.

that result from various c.m. H^+ velocities and (H_2^{+*}) orientations. The c.m. velocity circle exaggerated in Fig. 3 and given by v(max) represents the maximum velocity of H^+ in the H_2^+ c.m. frame. This velocity corresponds to a c.m. energy of about 5 eV above the $H^+ + H(2p)$ dissociation limit and results from a vertical Franck-Condon transition to the $2p\pi_u$ state at an internuclear separation of $1.5a_0$ as indicated in Fig. 4. We have chosen $1.5a_0$ because this value corresponds to the classical turning point of the $v=3$ vibrational state, the state with maximum population probability as determined by the Franck-Condon principle in H_2^+ formation from H_2.[6] From Fig. 3(c), we see that a 5-eV c.m. H^+ energy corresponds to a maximum laboratory scattering angle of 3.2°, with the H_2^+ internuclear axis perpendicular to the initial ion beam direction. It is at this angle (internuclear orientation) we ob-

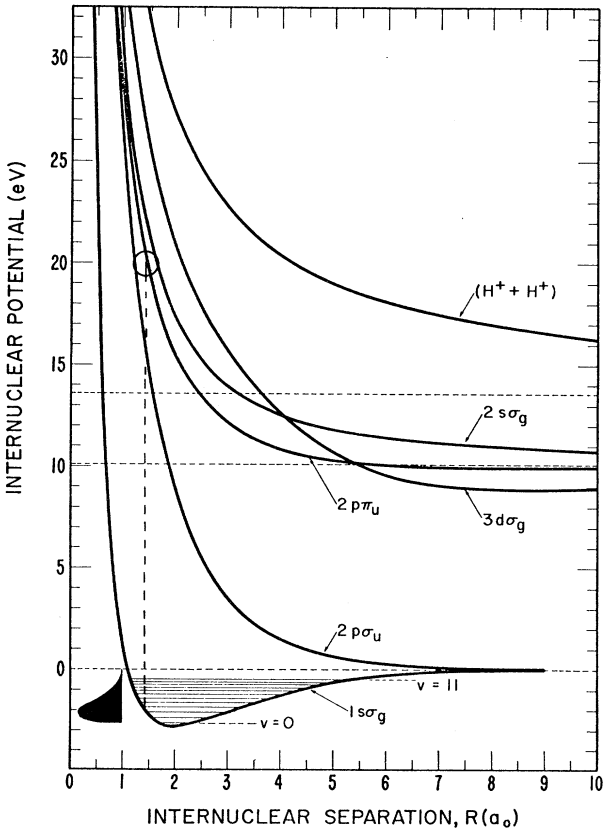

FIG. 4. Relevent H_2^+ potential curves. For collisions in which the H_2^+ internuclear-axis orientation is perpendicular to the beam direction, the H_2^+ molecular ion is excited by a Franck-Condon type transition to the $2p\pi_u$ state at an internuclear separation of 1.5 a.u. The maximum alignment of the H($2p$) results from the subsequent dissociation of $H_2^+(2p\pi_u)$.

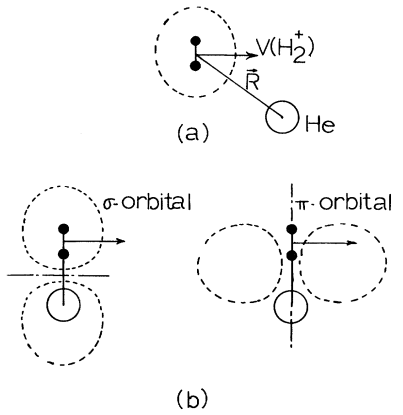

FIG. 5. (a) $1s\sigma_g$ state of the incoming H_2^+. (b) Near distance of closest approach, the nodal planes of σ-like (parallel to the beam direction) and π-like (perpendicular to the beam direction) orbitals.

serve maximum alignment of the H($2p$) resulting from the dissociation of $(H_2^+)^*$.

With the aid of the quasidiatomic MO correlation diagram and the polarization intensity measurements $I(\beta)$, we can obtain insights into the excitation mechanisms in H_2^+-He collisions. The polarization pattern $I(\beta)$ for small H^+ scattering angles, θ, represents intensity contributions from a sum of H_2^+ internuclear orientations and various c.m. H^+ velocities as illustrated in Fig. 3. By $\theta = \pm 3.25°$, only a small range of internuclear orientations and c.m. H^+ velocities contribute to the observed L_α-H^+ coincidence signal. Also at this angle we are observing $H(2p) + H^+$ from dissociating states of H_2^{+*} that are excited near the highest possible point of the repulsive potential curves.

Focusing our attention on the results at this angle, we note that the polarization intensity of the L_α radiation is that characteristic of $2p_{\pm 1} \rightarrow 1s$ transitions, using the H_2^+ internuclear axis as the quantization axis.[5] Thus, the observed final $H(2p_{\pm 1}) + H^+$ state results from the excitation and decay of the Π_u state of H_2^+, a result consistent with using 5 eV as the c.m. energy of H^+.

These results can be interpreted within the context of the quasidiatomic description of the excitation mechanism for H_2^+-He collisions. Figure 5 diagramatically illustrates the prediction of the MO diagram of Fig. 1, for the case of H^+ scattering at 3.25° (internuclear axis perpendicular to the beam axis). Rotational coupling, that is, $\sigma \rightarrow \pi$ transitions at the closest He-H_2^+ centers-of-mass distance, produces an excited π-like orbital

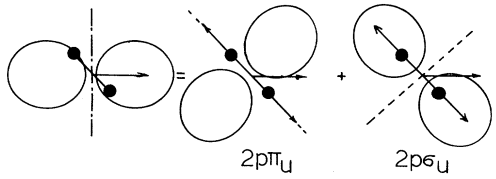

FIG. 6. $2p\pi_u$ and $2p\sigma_u$ wave functions for arbitrary internuclear orientation.

of the He-H_2^+ system, having for this orientation a nodal plane perpendicular to the beam axis. The wave function for the H_2^+-He system consists of a linear combination of the two wave functions having nodal character similar to that shown in Fig. 5. The excited-state wave function carried off by the H_2^+ is then π-like in character, with the H_2^+ internuclear axis in the nodal plane. Thus the final excited H_2^+ orbital has ungerade symmetry.

As the excited H_2^+ recedes from the He target the wave function remains frozen or locked into the H_2^+ internuclear axis producing a large amplitude for exciting the Π_u state. Being a Π_u state, the system dissociates to $H^+ + H(2p_{\pm 1})$ relative to the internuclear axis, that is, perpendicular to the beam direction, and provides the measured polarization distribution.

As shown in Fig. 6, other orientations α lead to excitation of other states, since the space-fixed Π-like wave function can be written as a coherent linear combination of body-fixed Π and σ wave functions.

Thus the two dominant excitation channels should be to the $2p\pi_u$ and $2p\sigma_u$ states. At an H_2^+ orientation perpendicular to the beam direction the excitation should be to the $2p\pi_u$ state, while for a parallel orientation the $2p\sigma_u$ state should dominate.

Thus the collision model here addresses a rather long outstanding problem in the understanding of molecular collision excitation and decay processes.[7] The problem was one of trying to interpret and reconcile the proton production in detailed Astron-band measurements with the fact that cross sections for L_α production in H_2^+-He collisions[7-10] are of the same order of magnitude. The results here clearly suggest that both states play a role and the excitation should be of comparable magnitude.

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Precise Measurements of Hyperfine Structure in the $2\,^3P$ State of ^{3}He

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Transitions within the $2\,^3P$ state of ^{3}He were measured to determine the parameters C, D, and E which represent respectively the contact, dipole, and tensor terms in the hyperfine Hamiltonian. For the first time, the effect of core polarization on the hyperfine structure has been detected. The present results are $C=-4283.84(1)$ MHz, $D=-28.02(6)$ MHz, and $E=+7.08(2)$ MHz and are consistent with the theoretical values after the combined fine and hyperfine interactions with the $2\,^1P$ state are taken into account in the analysis of the data.

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The spectra of excited states ($1snl$) of ^{3}He have been the subject of several recent experimental investigations. The techniques employed are level crossings and anticrossings,[1] various forms of Doppler-free laser spectroscopy,[2,3] and quantum beats in beam-foil spectra.[4] States of ^{3}He which have been studied include $n\,^3S$, $n\,^3D$, and $n\,^1D$ up to $n=6$, and $n\,^3P$ up to $n=8$. The measured intervals within these states are now known with accuracies ranging from 0.1 to 20 MHz. As the principal quantum number n increases, the ^{3}He$^+$ core dominates first the fine structure, then the exchange interaction, and ultimately (above $n\sim100$) the gross level structure, leading to two ionization limits. We hope to explore these effects to a precision of about 0.03 MHz using an extension of the optical microwave technique reported here.

In this Letter we report a measurement of the $1s2p\,^3P$ hyperfine structure which is 2 to 3 orders of magnitude more precise than earlier data. In particular we have measured with high precision the contribution of the p electron to the hyperfine structure, a phenomenon which was previously barely resolved from the noise,[3] and we have measured previously undetected core-polarization effects.

The hyperfine Hamiltonian is given by[5]

$$\mathcal{H}_{\rm hfs}=-2\mu_0\sum_{i=1}^{2}\left\{-\frac{8\pi}{3}(\vec{s}_i\cdot\vec{\mu})\delta(\vec{r}_i)-\frac{1}{r_i^{\,3}}\vec{l}_i\cdot\vec{\mu}+\frac{1}{r_i^{\,3}}\left[\vec{s}_i\cdot\vec{\mu}-\frac{3(\vec{s}_i\cdot\vec{r}_i)(\vec{\mu}\cdot\vec{r}_i)}{r_i^{\,2}}\right]\right\}. \quad (1)$$

The sum is taken over the two electrons, $\vec{\mu}$ is the nuclear magnetic moment, and $\vec{s}_i$, $\vec{l}_i$, and $\vec{r}_i$ have the usual meanings for an individual electron.

Within a pure ^3P state, Eq. (1) may be written

$$\mathcal{H}_{\rm hfs}=C\vec{I}\cdot\vec{S}+D\vec{I}\cdot\vec{L}+(2\sqrt{10})E\vec{I}\cdot(\vec{S}C^{(2)})^1, \quad (2)$$