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$He(3\,^3P)$ excitation in 1.5- and 3.0-keV $He^+ + H_2$ collisions

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Photon–scattered-atom coincidence measurements, for the collision $He^+ + H_2 \rightarrow He(3\,^3P) + H_2^+$, both with and without linear polarization analysis, have been made for small-angle scattering of He at 1.5 and 3.0 keV. A very large polarization for the $3\,^3P$ to $2\,^3S$ transition is observed at 1.5 keV, 1.5° and 3.0 keV, 1.25°. Also, the excitation probability exhibits small but definite structure as a function of scattering angle.

I. INTRODUCTION

Measurements by Eriksen and Jaecks[1] have demonstrated a large alignment of $He(3\,^3P)$ states excited in $He^+ + H_2$ charge transfer collisions. At certain incident energies and scattering angles, the alignment is even larger than for $He(3\,^3P)$ states excited in $He^+ + He$ collisions as measured by Vassilev *et al.*[2] and Jaecks *et al.*[3,4] Thus, the measurements reported in Ref. 1 indicate that surprisingly strong coherence effects can be observed in ion-molecule collisions.

The application of photon–scattered-particle coincidence techniques can help to determine the importance of a complete definition of the final state in an atomic-collision measurement. Measurements such as those by Eminyan *et al.*[5] for electron excitation of He, or those by Vassilev *et al.*[2] and Jaecks *et al.*,[3,4] are known to deal with pure states. On the other hand, the excited states observed by Berry *et al.*[6] for He^+ collisions with a foil are mixed states. In general, the states observed in photon-particle coincidence measurements of ion-molecule collisions will be mixed states, although Eriksen and Jaecks[1] have shown in one case that a nearly-pure-$He(3\,^3P)$ state was excited even though the final state was not completely defined.

Low-keV-energy $He^+ + H_2$ collisions in particular are of interest because work by Nathan and Isler[7] shows polarization of the H-α radiation. Using the results of Van Brunt and Zare[8] the polarization was interpreted to mean that the magnetic sublevels are unequally populated and the dissociation fragments are anisotropic. This interpretation is complicated by the fact that several molecular orbitals may contribute to the fluorescence, although McKnight and Gilliand[9] show the anisotropy of slow ions formed in other ion-molecule collisions.

Attempts at fully understanding the triatomic $He^+ + H_2$ interaction by total-cross-section measurements are complicated by the fact that one is averaging over a large number of collision variables. We have reduced the number of undetermined collision variables in the investigation of $He^+ + H_2 \rightarrow He(3\,^3P) + H_2^+$ by measuring the coincidence rate between 3889-Å radiation, with linear-polarization analysis, and scattered atoms at 1.5 and 3.0 keV for selected scattering angles. This paper is, in part, a more complete description of the work of Ref. 1, showing substantial linear polarization and nearly the maximum allowed theoretically at 1.5 keV, 1.5°.

We also report new measurements of the coincidence rate at 1.5 keV as a function of scattering angle, but without linear-polarization analysis. We interpret this rate to be proportional to the $He(3\,^3P)$ differential-excitation probability. The threshold of this probability gives information about the maximum "impact parameter" allowable in the excitation, and structure in the probability is of interest for two reasons. It may be caused by crossing of potential surfaces, and the energy and angular dependence of the structure can verify any scaling laws which apply to ion-molecule collisions.[10]

Those aspects of the apparatus and the experimental method which were not presented in Refs. 1 and 4 are described in Sec. II. Our results are given in Sec. III and discussed in Sec. IV.

II. APPARATUS

Referring to Fig. 1, a beam of He^+ ions is momentum analyzed by a magnet and directed toward the collision region at 0 where it interacts with a thermal beam of H_2 molecules. Perpendicular to the collision plane, an optics system collects radiation from 0 and selects it according to its wavelength and linear polarization. The axis of maximum transmission of the analyzer makes an angle β with respect to the incident-beam direction. Particles scattered to an angle θ and passing through slit S3 are detected by a conventional electron multiplier. A potential difference between the plates of the parallel-plate electrostatic analyzer allows only neutral particles to pass through S3.

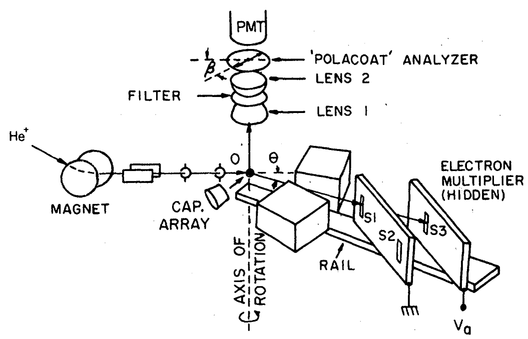

FIG. 1. Schematic diagram of the apparatus.

Pulses from the photomultiplier are detected in delayed coincidence with pulses from the electron multiplier.

In order to insure single-collision conditions, all data were taken at a sufficiently low scattering pressure that the neutral scattered particle count rate and photon count rate were linear functions of the scattering chamber pressure. As previously reported,[4] changes in scattering pressure did not change the results of our measurements of He($3\,^3P$) sublevel populations excited in $He^+ + He$ collisions.

There is a possibility of contributions to the photon signal from the Balmer-ζ line. The interference filter has a center wavelength of 3886 Å and a full width at half maximum (FWHM) of 36 Å so radiation from the $n = 8$ to $n = 2$ states of hydrogen atoms, $\lambda = 3888$ Å, would be accepted by the filter. Measurements by Isler and Nathan[7] suggest that this effect is less than 10%. At an incident energy of 700 eV, Isler and Nathan report a cross section for H-β emission of about 7×10^{-19} cm^2. The trend of the Balmer lines from their Fig. 2 indicates that H-ζ is at least a factor of 20 smaller than the H-β cross section. They report the He($3\,^3P$) excitation cross section to be 2.8×10^{-19} cm^2; therefore, the contribution of H-ζ to our signal is at most 12%, and probably much less since the cross section for He($3\,^3P$) excitation is rising from 0.7 to 1.5 keV (see Ref. 11).

Also, our coincidence technique discriminates against cascade contributions. We estimate this effect to be less than 5% of our signal, although in a standard total-cross-section measurement the cascade contribution can be as high as 20% for $He^+ + He$ collisions.[12] A time resolution of 100 ns is used since the lifetime of the He($3\,^3P$) state is 110 ns. Cascading states have lifetimes of 150 ns or more.[13]

III. DATA

Figure 2 shows polar plots of the real coincidence rate versus the angle β at several combinations of incident laboratory kinetic energy and laboratory scattering angle. For a given energy and scattering angle, data were taken at polarizer angles of 0°, 45°, 90°, and 135° with a final datum point being the average of at least two 24-h determinations of the count rate. For each pattern the z axis is the direction of the He^+ beam and scattered neutral particles are detected above the z axis. Polarization patterns taken at 1.5 keV employed a new delay for the coincidence electronics which was determined empirically. The peculiar angle 2.33° was chosen to give a value of $T_0\theta^2$ of 8 keV deg^2. Below this value, it has been shown by Bray *et al.*[10] that little vibrorotational excitation occurs for the direct scattering process

$$He^+ + H_2 \rightarrow He^+ + H_2(\text{grnd. electronic st.})\,. \qquad (1)$$

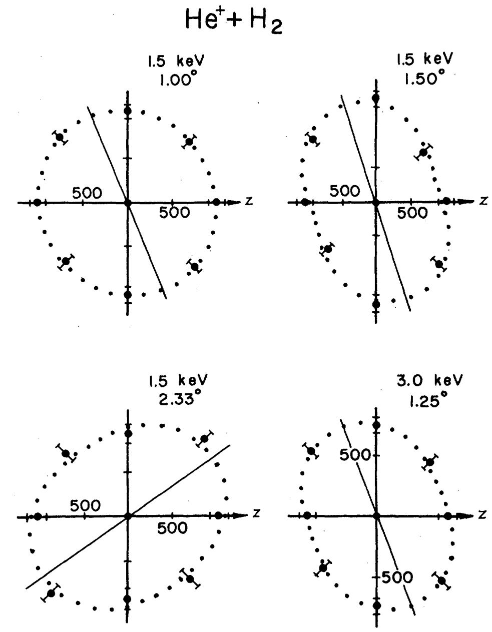

FIG. 2. Polar plots of the average number of real coincidences per 10^9 scattered neutral particles vs β for three scattering angles at 1.5 keV, and one angle at 3.0 keV. The data are shown by solid dots with error bars and the dots without error bars are best fits to the data as described in Ref. 4. In all the plots, the incident beam direction is along the z axis (to the right in the figure) and the neutral particles are detected above the z axis.

Figure 3 shows a graph of the coincidence rate per scattered neutral particle with no linear-polarization analysis. Each datum point is the average of usually two or more 24-h determinations of the count rate. For comparison, Fig. 4 shows similar data for $He^+ + He$ collisions at 3.0 keV. Error bars represent estimates of one-standard-deviation random counting error.

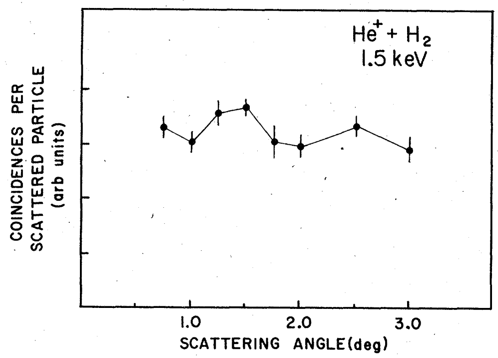

FIG. 3. Plot of the number of coincidences per 10^6 scattered neutral particles, with no polarization analysis, as a function of laboratory scattering angle for $He^+ + H_2$ collisions.

The error for each determination of the count rate σ_i is obtained as described in Ref. 4, and the total error σ_T at a particular scattering angle was obtained from

$$\sigma_T^{-2} = \sum_{i=1}^{N} \sigma_i^{-2} , \qquad (2)$$

where N is the number of determinations at a particular β and θ, and σ_i is the error for each determination.

IV. DISCUSSION

A. Measurements with polarization analysis

We have interpreted the polarization patterns of Fig. 2 using the results of Fano and Macek.[14] The source of 3889-Å radiation is characterized by three alignment parameters ($A_0'^{\rm col}$, $A_{1+}'^{\rm col}$, and $A_{2+}'^{\rm col}$) and one orientation parameter ($O_{1-}'^{\rm col}$), although our measurements do not determine $O_{1-}'^{\rm col}$ since the light is not analyzed according to its circular polarization. As described in Ref. 1, the intensity of the emitted radiation is given by

$$I(\beta) = \tfrac{1}{3} CS \{1 + \tfrac{1}{4} h^{(2)} \overline{G}^{(2)} [A_0'^{\rm col} + 3A_{2+}'^{\rm col} + 3(A_0'^{\rm col} - A_{2+}'^{\rm col}) \times \cos 2(\beta - \alpha)]\} , \qquad (3)$$

where β is the angle between the axis of maximum transmission of the linear polarizer and the incident-beam direction, α is the angle between the maximum of the polarization pattern and the incident-beam direction, and C and S are constants. The factor $h^{(2)} = -2$, and the factor $\overline{G}^{(2)}$, which accounts for the depolarization of the light owing to the fine structure of the He($3\,^3P$) level, is a time average of an expression given by Macek[15]; $\overline{G}^{(2)}$ is

$$\overline{G}^{(2)} = \frac{1}{3} \sum_{J=0,1,2} (2J+1)^2 \begin{Bmatrix} J & J & 2 \\ 1 & 1 & 1 \end{Bmatrix}^2 . \qquad (4)$$

For the case of He($3\,^3P$) excitation, $\overline{G}^{(2)} = \frac{5}{18}$. Equation (3), in general, describes an hourglass-shaped pattern on a polar plot of I vs β, there being three undetermined parameters $A_0'^{\rm col}$, $A_{2+}'^{\rm col}$, and α. We have determined least-square best fits of Eq. (3) to our data as described in Ref. 4, these fits being shown in Fig. 2 by dotted lines. The major axis of each fit is shown by a solid line.

Of special interest here is the ratio, R, of the minimum intensity $I_{\rm min}$ to the maximum $I_{\rm max}$ for a given pattern. From Eq. (3) one finds

$$R = \frac{I_{\rm min}}{I_{\rm max}} = \frac{1 - \overline{G}^{(2)}(3A_{2+}'^{\rm col} - A_0'^{\rm col})}{1 - 2\overline{G}^{(2)} A_0'^{\rm col}} . \qquad (5)$$

The values $A_{2+}'^{\rm col}$ and $A_0'^{\rm col}$ obey the inequality relations

$$-1 \le \overline{A}_0'^{\rm col} \le 0.5, \quad -1 \le A_0'^{\rm col} \le 0.5 , \qquad (6)$$

where $\overline{A}_0'^{\rm col} = \frac{1}{2}(3A_{2+}'^{\rm col} - A_0'^{\rm col})$. As a function of $A_{2+}'^{\rm col}$ and $A_0'^{\rm col}$, R has a minimum value of 0.46 when $A_0'^{\rm col} = -1$ and $A_2'^{\rm col} = 0$. A state characterized by these parameters would be a pure state with $m_l = 0$ relative to the z' axis, which lies along the direction of the maximum in the polarization pattern.

It is interesting to note that without the fine-structure depolarization, $\overline{G}^{(2)} = 1$ and the minimum R is zero. Under these conditions, the observation of a minimum R only implies that $3A_{2+}'^{\rm col} - A_0'^{\rm col} = 1$, a less restrictive conclusion than can be made with the fine-structure depolarization present.

Experimental values of R were obtained from the computer fits for each pattern in Fig. 2. Table I summarizes these values as well as values of R for data for He⁺+He at 3.0 keV.[3,4] We also include results for He⁺+He at 1.5 keV, some re-

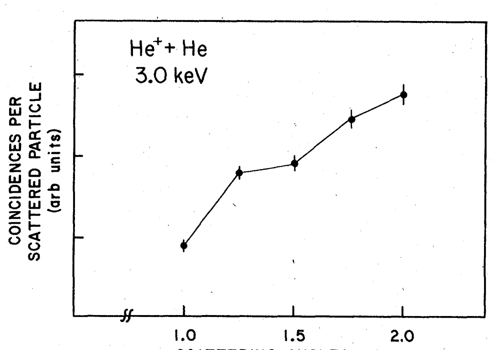

FIG. 4. Data equivalent to those in Fig. 3 only for $He^+ + He$ collisions. A plot of the number of coincidences per 10^6 scattered particles, with no polarization analysis, calculated from data in Ref. 4 as described in the text.

TABLE I. Measured and corrected values of the ratio R. T_0 and θ are the laboratory incident kinetic energy in keV and laboratory scattering angle in degrees, respectively. The correction of R_{meas} to R_{corr} is explained in the text.

T_0 (keV)	θ (deg)	R_{meas}	R_{corr}
He^+-H_2			
1.5	1.00	0.90±0.09	0.89±0.09
1.5	1.50	0.62±0.04	0.60±0.04
1.5	2.33	0.82±0.07	0.79±0.07
3.0	1.25	0.72±0.10	0.70±0.10
He^+-He			
1.5	1.25	0.75±0.11	0.73±0.11
1.5	1.50	0.72±0.14	0.70±0.14
3.0	1.00	0.72±0.09	0.70±0.09
3.0	1.25	0.72±0.10	0.70±0.10
3.0	1.50	0.84±0.09	0.83±0.09
3.0	1.75	0.79±0.09	0.78±0.09
3.0	2.00	0.82±0.11	0.81±0.11

TABLE II. Comparison of angles θ_{mom} and θ_{sym} as defined in the text. The inelastic energy loss is denoted ΔQ, and values of ΔQ were chosen as described in the text.

T_0 (keV)	θ (deg)	θ_{sym} (deg)	ΔQ (eV)	θ_{mom} (deg)
3.05	1.25	109 ± 11	13.9	97
		(maj)	14.9	98
			16.3	98
			29.5	104
1.52	1.00	112 ±10	13.9	105
		(maj)	14.9	106
			16.3	107
			29.5	118
1.52	1.50	107 ± 9	13.9	102
		(maj)	14.9	102
			16.3	103
			29.5	112
1.52	2.33	125 ±23	13.9	100
		(min)	14.9	100
			16.3	101
			29.5	107

ported[16] and some not. As described in Ref. 1, the values of R have been corrected to account for the fact that the polarization analyzer accepts some light with polarization perpendicular to its own axis. Also, a more careful analysis of the error in R is reported here. The errors stated in Table I are estimates of one standard deviation and are calculated using

$$\sigma_R^2 = \left(\frac{\partial R}{\partial I_{max}}\right)^2 \sigma_{I_{max}}^2 + \left(\frac{\partial R}{\partial I_{min}}\right)^2 \sigma_{I_{min}}^2 , \quad (7)$$

where R is defined in Eq. (5), σ_R is the standard deviation in R, and $\sigma_{I_{max}}$ and $\sigma_{I_{min}}$ are obtained from the computer fits as described in Ref. 4.

The data in Table I are interesting in several respects. First, the $He^+ + H_2$ data at 1.5 keV, 1.50° and 3.0 keV, 1.25° show at least as much polarization as any of the data for He^+ + He studied so far. Second, the ratio R at 1.5 keV, 1.5° for $He^+ + H_2$ is 0.60 ±0.05, sufficiently close to the minimum value of 0.46 to suggest the excitation of a nearly-pure-He(3^3P) state. We feel these results are surprising in view of the substantial difference between He and H_2 targets. It is well known that in He^+ + He collisions little excitation of the ionic atomic state occurs at these energies. Thus, the recoiling He^+ in a He^+ + He charge transfer collision is almost certainly in the 1s state. On the other hand, in $He^+ + H_2$ charge transfer collisions, the recoiling H_2^+ has many vibrorotational states available to it as well as many electronic states. Considering that the intensity of the radiation is summed over these final states, it is surprising that the polarization in $He^+ + H_2$ collisions is not weak at every scattering angle.

The direction of the symmetry axes of the polarization patterns in Fig. 2 suggests that the momentum transfer axis is important in the $He^+ + H_2$ collisions we study. Listed in Table II, for each incident energy and scattering angle studied, are θ_{sym}, the angle of the major or minor axis of each polarization pattern, and θ_{mom}, the angle of the momentum transfer axis as calculated for selected inelastic energy losses ΔQ. Both angles θ_{sym} and θ_{mom} are defined relative to the incident-beam direction. The errors reported for θ_{sym} are estimates of one standard deviation, and are obtained using Eq. (12) of Ref. 4.

In order to calculate the angle θ_{mom}, we assumed that the H_2^+ molecule did not dissociate immediately but recoiled as a single particle; that is, in the equations for the conservation of energy and momentum we chose the mass of the recoiling particle to be twice the proton's mass. This assumption should be valid in light of the recent measurements by Bray *et al.*[10] which suggests that there is little vibrational excitation for values of the parameter $T_0\theta^2$ less than 8 keV deg^2. Although it makes a difference of only a few percent, we have taken into account the mass of the electron which is transferred from the target to the projectile.

It was also necessary to assume a value for the inelastic energy loss ΔQ. In Ref. 1 we assumed several arbitrary values for ΔQ, but have now recalculated θ_{mom} for more relevant energy losses. Consider the process

$$He^+(1s) + H_2(X\,^1\Sigma_g; \nu=0) \to He^*(3\,^3P) + H_2^+(n'; \nu'), \quad (8)$$

where n' and ν' denote the final electronic and vibrational state of the H_2^+ molecule. Since the ionization potential of He is about 24.5 eV, the excitation energy of He($3\,^3P$) is about 23.0 eV, and since the ionization potential of H_2 is about 15.4 eV, the minimum inelastic energy loss is 13.9 eV. If the Franck-Condon principle is obeyed, the most probable transitions are to the $\nu'=0$ and the $\nu'=4$ vibrational states of the $H_2^+(1s\sigma_g)$ state, implying energy losses of 13.9 and 14.9 eV, respectively. To excite the highest bound vibrational level of the H_2^+ ground electronic state would require an energy loss of about 16.3 eV. If electronic excitation of the H_2^+ occurred to the $2p\sigma_u$ level, the Franck-Condon principle would imply an energy loss of at least 29.5 eV. Therefore, our calculations of θ_{mom} were made for energy losses of 13.9, 14.9, 16.3, and 29.5 eV, covering the several lowest excitations.

It can be seen from Table II that the direction of the observed symmetry axes are all in agreement with the momentum transfer axes. Although the data do not extend over a large range of incident energy and scattering angle, there is some indication that the momentum transfer direction is important. It is known that theories such as the Born approximation require the emitted radiation to be symmetric about the momentum transfer axis.

The Born approximation has been used to analyze dissociation processes in H_2^+ + He collisions. Theoretical work has been done by Green and Peek[17] and measurements on the dissociation fragments have been made by Gibson *et al.*[18] and Sauers *et al.*[19] Also, Isler[20] has reported polarization measurements of the fluorescence in total cross sections. None of the work using the Born approximation, however, is directed toward a calculation of the He radiation emitted in charge exchange processes, and so this work is only related to ours in a very general way. Most importantly, however, the validity of the Born approximation is very suspect for collisions where there is likely to be a large distortion of the electron wave functions. See, for example, comments by Isler[20], Gibson *et al.*,[18] and Green and Peek.[17]

B. Measurements without polarization analysis

Figure 3 shows the coincidence rate per scattered neutral particle as a function of scattering angle, with no polarization analysis. We have interpreted this rate as proportional to the differential excitation probability, although our measurements do not detect radiation from states with transition moment along the viewing axis. In similar measurements for He^+ + He, the symmetry precludes the formation of these states. With an H_2 molecular target, this symmetry is broken and such moments can be formed.

The differential probability is a generally constant function of the scattering angle with a small but definite peak near 1.5°. For comparison, we have included in Fig. 4 the same kind of measurement for He^+ + He collisions. In this case the probability is a much stronger function of scattering angle, and exhibits a slight minimum near 1.5.

The data of Fig. 3 set an upper limit on the threshold for the excitation of He($3\,^3P$), namely, $\tau_{threshold} \leq 1.1$ keV deg, where τ is measured in center-of-mass units. The threshold observed[21] for He($3\,^3P$) excitation in He^+ + He collisions is 1.5 keV deg, indicating a less violent collision at threshold for $He^+ + H_2$.

Structure in angular differential cross section in ion-atom collisions is often explained by interference between two molecular states. For example, Lorents *et al.*[21] have observed regular oscillations in the differential cross section for He($2\,^3S$) excitation in He^+ + He collisions, and Rahmat *et al.*[22] have observed similar structure in the excitation of He($3\,^3P$) states. Typically the wavelengths for such structure are 0.6 to 0.7 keV deg. If the structure in Fig. 3 is a part of a series of regular oscillations, its wavelength would be approximately 1.1 keV deg, in qualitative agreement with data from ion-atom collisions.

Finally, the structure shown in Fig. 3 will be valuable in checking the validity of any scaling laws applicable to ion-molecule collisions. Bray *et al.*[10] have shown that at least one differential cross section for direct excitation in $He^+ + H_2$ collisions scales with the reduced parameter τ. This parameter is known to be important in ion-atom collisions, but the extent of its applicability to ion-molecule collisions is unknown. The energy and angular dependence of the structure can help resolve this question.

V. SUMMARY

In summary, we have shown that there is a large linear polarization of the 3889-Å radiation emitted in small-angle $He^+ + H_2$ charge exchange collisions, and that at least at some energies and scattering angles the radiation is characteristic of a nearly pure state. Thus, the H_2 molecular target appears to be sufficiently simple that atomic-target-like coherence effects occur.

In previously reported work for $He^+ + H_2$, experimental efforts have been primarily in one of three areas: measurements of the wavelength and intensity of emitted light, measurements of the energy and angular dependence of slow ions, and measurements of the energy and angular dependence of fast scattered particles. Until this time, however, there have been few data correlating these different areas. In an ion-atom or atom-atom collision, it is usually sufficient to know the inelastic energy loss and the scattering angle in order to infer the excitations of the collision partners. However, due to the rich structure of molecules, knowledge of these two parameters is not always sufficient to identify the electronic states in a several-keV ion-molecule collision. Thus, future experiments should be designed to provide information about the molecular states simultaneously with the usual information about the scattering angle and energy of the fast scattered atoms. This information can only be determined by an appropriate coincidence technique, for example, by detecting radiation from an excited atomic fragment in coincidence with a fast scattered ion or atom with energy analysis of the fast scattered particle. One could then infer the final excited state of the molcule, as well as the state of the scattered particle, in collisions where the energy and angle of the scattered particle are defined.

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