Polarization studies of H(2*p*) charge-exchange excitation: H^+-Ar collisions

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Alignment and orientation for H(2p) excitation in H^+-Ar collisions have been measured for incident proton energies of 1.5 and 3 keV, and in the range of scattering angles between 0.5° and 3.5°. From the experimental results the relative population of magnetic substates H($2p_0$) and H($2p_{\pm 1}$) is extracted. The observed magnitude of the H($2p_0$) excitation suggests that, in addition to a $3d\sigma$-$3d\pi$ rotational coupling, a $3d\sigma$-$4p\sigma$ radial coupling is effective.

I. INTRODUCTION

Angular correlation and polarization studies involving collisionally excited characteristic photons and Auger electrons have found increasing interest in the past years. Most of these studies have been devoted to outer-shell excitation and inner-shell ionization processes in electron-atom and ion-atom collisions (see, e.g., Refs. 1–3). Recently, such investigations were extended to charge-changing processes (see, e.g., Refs. 4–6). Measurements of this type provide detailed information about the underlying collision mechanism; in particular, they allow one to derive magnitude and relative phase of scattering amplitudes.[2] More formally, this information may be expressed in terms of multipole moments, commonly referred to as orientation and alignment.[7] These multipoles are related to the spatial distribution of atomic angular momenta, and thus reflect the anisotropy of the atomic ensemble.

We are at present performing a detailed investigation of hydrogen $2p$ excitation in charge-changing collisions,

$$H^+ + X \rightarrow H(2p) + X^+ \ . \tag{1}$$

A polarization analysis, or an angular distribution measurement of Lyman-α photons emitted during the decay of the H(2p) state (mean lifetime 1.6 ns) to the H(1s) ground state yields information about the collisionally excited H(2p) subensemble. In this paper we report on the study of the H^+-Ar collision system. From these measurements, we were able to determine the relative orientation vector and the components of the alignment tensor for impact energies of 1.5 and 3 keV, and in the range of projectile scattering angles between about 0.5° and 3.5°.

In order to describe the collision process one may introduce scattering amplitudes $f(m_1,m_2)$, with m_1 and m_2 referring to the z component of the orbital angular momentum of H(2p) and Ar^+, respectively. Here we use a right-handed coordinate frame with the z axis along the incident projectile's direction, the outgoing projectile's direction lying in the x-z (scattering) plane, and the y axis perpendicular to the scattering plane. One then obtains the monopole $\langle T(1)_{00}\rangle$, as well as the nonzero, spin-averaged components of the orientation vector $\langle T(1)_{11}\rangle$ and of the alignment tensor $\langle T(1)_{2Q}\rangle$ ($Q=0,1,2$) as

$$\langle T(1)_{00}\rangle = \sigma(2p)/\sqrt{3} \ ,$$

$$\langle T(1)_{11}\rangle = -i\sqrt{2}\,\mathrm{Im}\left[\sum_{m_2} f(1,m_2)f(0,m_2)^*\right] ,$$

$$\langle T(1)_{20}\rangle = (\sqrt{2}/\sqrt{3})\sum_{m_2}[\,|f(1,m_2)|^2 - |f(0,m_2)|^2\,] ,$$

$$\langle T(1)_{21}\rangle = -\sqrt{2}\,\mathrm{Re}\left[\sum_{m_2} f(1,m_2)f(0,m_2)^*\right] , \tag{2}$$

$$\langle T(1)_{22}\rangle = \sum_{m_2} f(1,m_2)f(-1,m_2)^*$$

with

$$\sigma(2p) = \sum_{m_1}\sigma_{m_1}$$

the differential scattering cross section for H(2p) charge-exchange excitation, and

$$\sigma_{m_1} = \sum_{m_2} |f(m_1,m_2)|^2$$

the corresponding partial cross sections. In the present investigation the final state of the remaining Ar^+ is not specified; therefore we have to sum over m_2. As we shall see later on, this (incoherent) summation over m_2 leads to a depolarization of the observed Lyman-α radiation. In deriving Eqs. (2), use was made of the reflection invariance in the scattering plane requiring

$$f(m_1,m_2) = (-1)^{m_1+m_2} f(-m_1,-m_2) \ . \tag{3}$$

In contrast to a recent measurement on the same collision system[5] where the alignment tensor components were derived from the angular distribution of photons emitted in coincidence with scattered projectiles, we have now performed a full polarization analysis of the emitted photons, thereby additionally obtaining the orientation vector. The degrees of linear and circular polarization (Stokes parameters[8]) are given by

$$P_1 = [I(0°) - I(90°)]/[I(0°) + I(90°)] \ ,$$

$$P_2 = [I(45°) - I(135°)]/[I(45°) + I(135°)] \ , \tag{4}$$

$$P_3 = [I(-) - I(+)]/[I(+) + I(-)] \ .$$

$I(\beta)$ is the intensity transmitted by a linear polarizer, with β the angle between the plane of polarization and the incident projectile axis (z axis). $I(+)$ and $I(-)$ denote the intensity of circularly polarized light with positive and negative helicity, respectively. Alignment and orientation are obtained from a measurement of the Stokes parameters perpendicular to the z axis using[2]

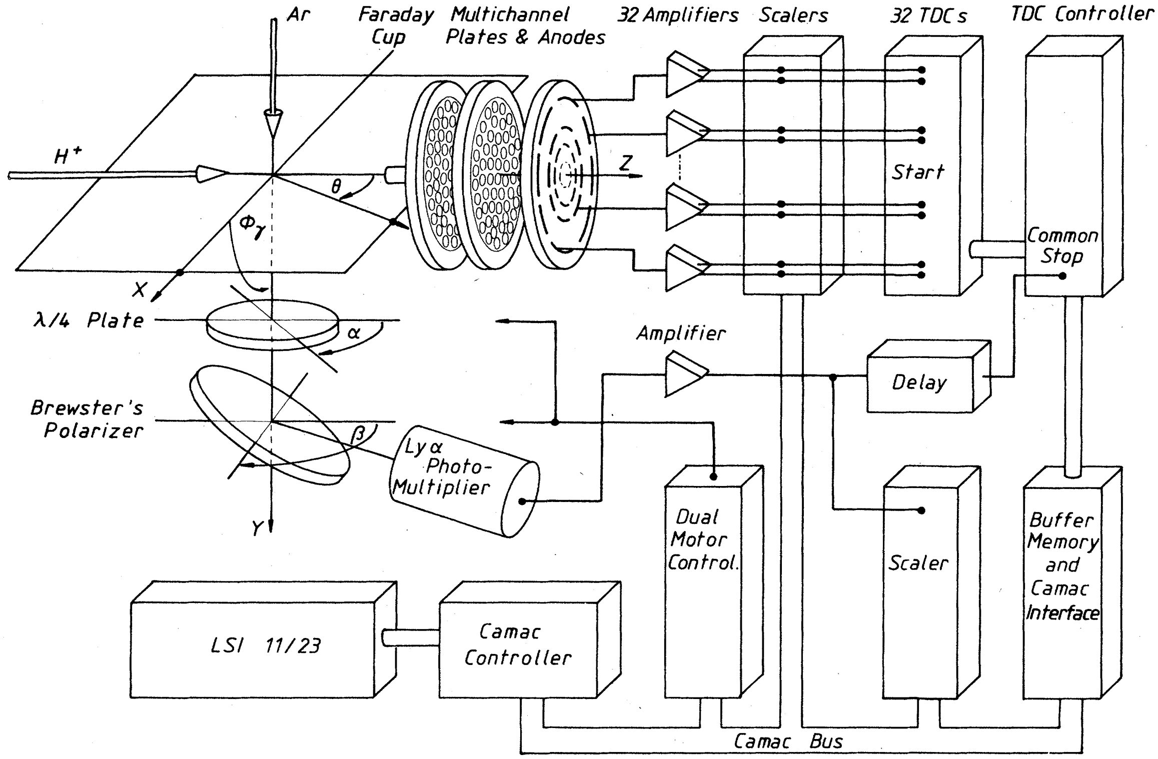

FIG. 1. Experimental setup (schematic).

$$P_1 = -[\langle T(1)_{22}\rangle \cos(2\phi) + (\sqrt{3}/\sqrt{2})\langle T(1)_{20}\rangle]/I ,$$
$$P_2 = 2\langle T(1)_{21}\rangle \sin(\phi)/I ,$$
$$P_3 = \tfrac{14}{3} i \langle T(1)_{11}\rangle \sin(\phi)/I , \quad (5)$$
$$I = 2\sqrt{3}\langle T(1)_{00}\rangle + \langle T(1)_{22}\rangle \cos(2\phi) - (1/\sqrt{6})\langle T(1)_{20}\rangle ,$$

where ϕ is the azimuthal angle of the photon detection relative to the scattering plane defined by the incoming and outgoing projectile's directions. In Eqs. (5) the depolarization caused by the spin-orbit interaction is taken into account.[2] The hyperfine interaction is weak in hydrogen and was neglected. It follows immediately from the above equations that only P_1 can be different from zero in measurements where ϕ is not specified (for instance, if the outgoing projectile is not detected). The total polarization P, corrected for the depolarization caused by the fine-structure coupling, is given by[9]

$$P^2 = (\tfrac{7}{3})^2 P_1^2 + (\tfrac{7}{3})^2 P_2^2 + P_3^2 . \quad (6)$$

This total polarization P provides a check for the coherence of the excited states: In case of complete coherence we have $P = 1$ (Ref. 9).

II. EXPERIMENTAL METHOD

The experimental arrangement consists of an ion gun, a scattering chamber with a polarization-sensitive detector for Lyman-α radiation, a position-sensitive detector for scattered (fast) hydrogen atoms, and the coincidence electronics (Fig. 1). Some details of this apparatus have been described previously.[5]

Ions with kinetic energies of a few keV are produced by a duoplasmatron ion source, and mass-analyzed in a 30° magnet. The ion beam enters the scattering chamber through a decelerator-lens system and suitably chosen diaphragms reducing its diameter to about 2 mm. 25 mm beyond the last diaphragm the beam hits a thermal gas target and is collected about 300—1200 mm (depending on the scattering angles chosen) downstream in two concentrical Faraday cups having diameters of 3 and 6 mm, respectively. Typically, more than 95% of the beam intensity is collected in the inner cup.

Photons produced in the collision region are detected with a photomultiplier (EMR 542J) having a LiF window and a KBr photocathode. This photomultiplier has a short-wavelength cutoff at 105 nm; its long-wavelength limit is about 155 nm. The photomultiplier detects the interaction region through a polarization-sensitive device. This consists of a suitably chosen mirror arranged at Brewster's angle[10] (about 60° for the mirror materials of interest here) with respect to the photon direction. This arrangement effectively transmits light with an electric vector perpendicular to the plane formed by the incident and the reflected light directions; it strongly suppresses the in-plane component. This linear-polarization analyzer detects the collision region at $\theta_\gamma = 90°$ with respect to the incident ion beam; it is mounted on a turntable to allow rotation about its optical axis as is necessary for polarization measurements. Two different reflecting mirrors were available. One is a plane LiF mirror which shows a large instrumental polarization of 90%. This was tested with the help of a second polarizer of the same type; as a light source we used an ion gauge operated with hydrogen gas together with a LiF-O_2 filter.[10–12] The other material was a curved Suprasil mirror of 6 cm radius which shows an instrumental polarization of 65%. This lower polarization was compensated by the larger efficiency of this mirror which was about 5 times as large as compared to the LiF mirror.

For the circular polarization measurements a quarter-wave plate made of MgF_2 was used. This $\lambda/4$ plate was tested in a series of measurements (compare, e.g., Ref. 13) using the above-mentioned linear polarizer-analyzer combination with LiF mirrors.[14] It was found that the $\lambda/4$ plate was slightly imperfect, showing an instrumental (circular) polarization of 99%. All measurements reported below have been corrected accordingly.

Fast neutralized projectiles scattered through selected scattering angles are detected in a position-sensitive detector consisting of two microchannel plates and an anode array. The anode array essentially consists of 32 individual electrodes arranged in such a way as to cover eight azimuthal angles ϕ for each of four scattering angles θ_s. Pulses from the 32 anodes are processed separately. With the help of two 16-fold amplifier-discriminator cards they are used as start inputs for 32 time-to-digital converters (TDC, LeCroy System 4290). Pulses from the Lyman-α photomultiplier are suitably delayed and served as stop inputs. Typical time windows were 512 ns, with a time resolution of about 20 ns. The experiment is automatically controlled by a LSI 11/23 microprocessor. Via a CAMAC dataway it positions the polarization analyzer and reads the time information from the 32 TDC's as well as the counting rates from the individual scalers. Typical counting rates were up to 10^5/s per individual scattered projectile channel, and 10/s in the photon channel. The time necessary to obtain a full polarization data set (see, e.g., Fig. 3) was of the order of 1–3 weeks.

III. RESULTS

A. Integrated polarization measurements

In order to test some of the properties of the linear and circular polarization analyzers we have measured the noncoincident linear polarization of light emitted in proton–rare-gas collisions. In such integrated polarization measurements (i.e., integrated over all projectile-scattering angles) we have $P_2=P_3=0$ [cf. Eq. (4)], P_1 was measured by placing the linear polarizer at a fixed angle β and rotating the quarter-wave plate to different angles α. Measurements were performed at $\beta=0°$ as well as 90°, thereby providing an additional test of the properties of the $\lambda/4$ plate. The integrated linear polarization P_1 was obtained from a measurement at two angles $\alpha=0°$ and 45°, where α is the angle between the fast axis of the $\lambda/4$ plate and the incident proton direction:

$$P_1=\pm[I(\alpha=0°,\beta)-I(\alpha=45°,\beta)]/I(\alpha=45°,\beta), \quad (7)$$

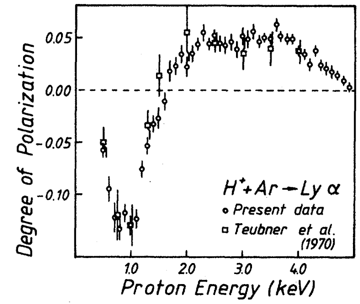

FIG. 2. Integrated polarization P_1 vs incident proton energy for H^+-Ar collisions.

where the + (−) sign corresponds to $\beta=0°$ (90°). Results of the integrated linear polarization measurements for incident proton energies between about 1 and 5 keV are given in Fig. 2. They are found to agree reasonably well with recent measurements of Teubner *et al.*[10]

B. Differential polarization measurements

In Fig. 3 a set of polarization measurements for $\theta_s=1.4°$ are presented. The data were measured simultaneously for the eight azimuthal angles ϕ shown. The orientation vector and the three components of the alignment tensor are obtained from such data by least-squares fits to Eqs. (5). The best fits are shown as solid lines. As can be seen, the circular polarization P_3, and hence the orientation vector, changes its sign when going from $\phi=90°$ to 270°.

In Fig. 4 the relative orientation vector $\langle T(1)_{11}\rangle/\langle T(1)_{00}\rangle$ as well as the three components of the alignment tensor $\langle T(1)_{2Q}\rangle/\langle T(1)_{00}\rangle$ $(Q=0,1,2)$ are given for an incident proton energy of 1.5 keV. Measurements are presented for scattering angles in the range of about 0.5°–3.5°, together with recent measurements by Hippler *et al.*[5] at somewhat larger scattering angles of up to 6°. In general, the earlier results obtained from a photon-scattered ion angular correlation measurement connect well to the present polarization measurements. In Fig. 5 data for an incident energy of 3 keV are presented, while Fig. 6 displays the total polarization P [Eq. (6)] measured perpendicular to the scattering plane. The full range of impact parameters b covered in the 1.5- and 3-keV measurements lies between 0.9 and 2.6 a.u. Transformation of scattering angles θ_s to impact parameters was done with the help of a Moliere potential (see, e.g., Ref. 15).

IV. DISCUSSION

There are some features of the presented alignment and orientation data which should be noted, namely,

(a) a small total polarization P of about 35%;

(b) a nonzero orientation, which is negative (positive) for 1.5 keV (3 keV) over most of the investigated range of scattering angles;

(c) the $\langle T(1)_{20}\rangle$ component of the alignment tensor has a minimum for impact parameters around 1.7 a.u.; in the 1.5-keV data it becomes positive for impact parameters

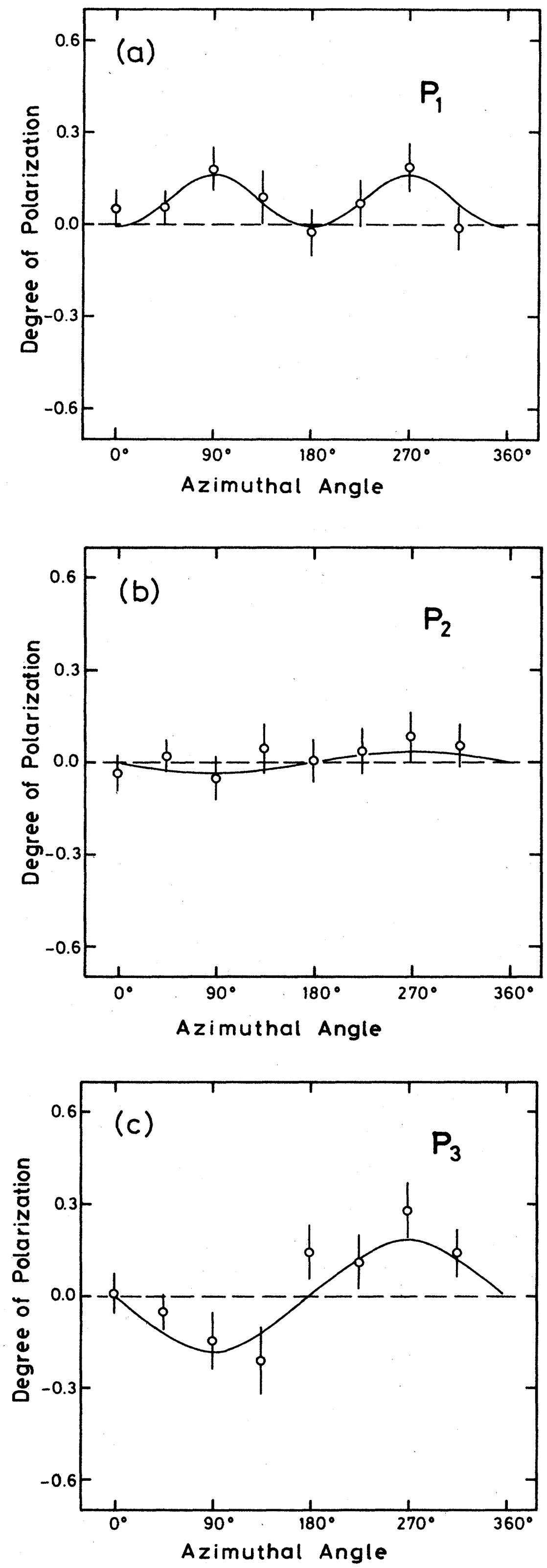

FIG. 3. Degree of polarization vs azimuthal angle ϕ for 1.5-keV H^+-Ar collisions and scattering angle $\theta_s = 1.4°$. The two linear (P_1, P_2) and the circular (P_3) polarizations are shown.

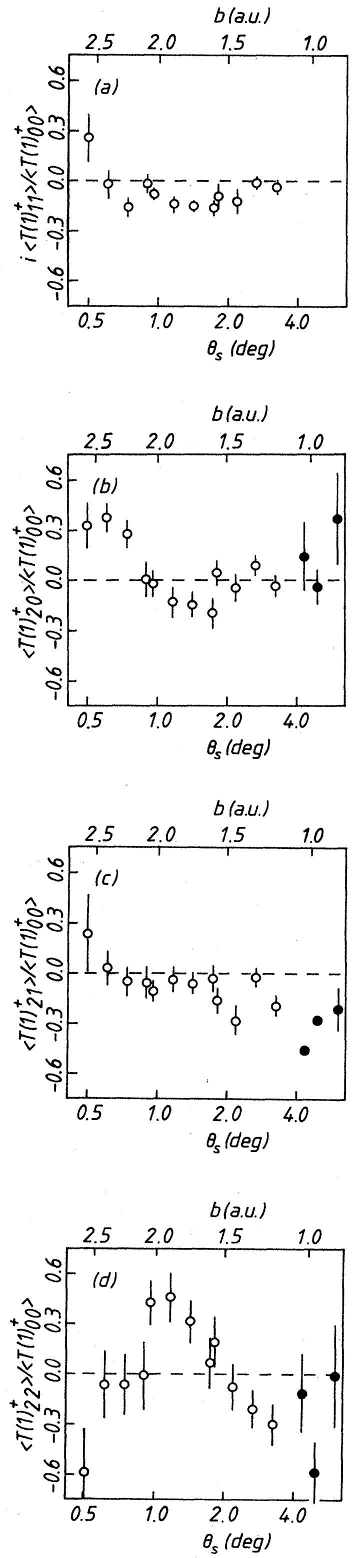

FIG. 4. Relative orientation vector $\langle T(1)_{11}\rangle / \langle T(1)_{00}\rangle$ and alignment tensor components $\langle T(1)_{2Q}\rangle / \langle T(1)_{00}\rangle$ ($Q = 0,1,2$) vs scattering angle θ_s for 1.5-keV H^+-Ar collisions. Solid circles are from Hippler *et al.*[5]

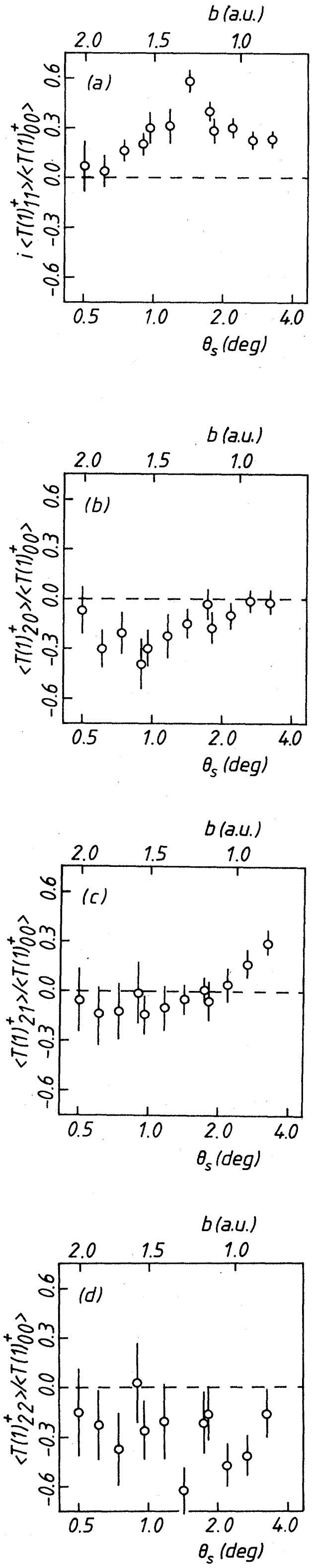

FIG. 5. Relative orientation vector $\langle T(1)_{11}\rangle/\langle T(1)_{00}\rangle$ and alignment tensor components $\langle T(1)_{2Q}\rangle/\langle T(1)_{00}\rangle$ ($Q=0,1,2$) vs scattering angle θ_s for 3-keV H^+-Ar collisions.

smaller than 1 a.u. and larger than 2 a.u.;

(d) in the 1.5-keV measurements the $\langle T(1)_{22}\rangle$ component shows a strong variation with impact parameter; this is not observed in the 3-keV data.

The features of the alignment tensor and orientation vector given above may provide some insight in the underlying excitation mechanism. In a quasimolecular picture[16,17] H(2p) excitation should take place in a two-step mechanism. In a first step, the

$$(1s\sigma^2 2s\sigma^2 2p\sigma^2 2p\pi^4 3s\sigma^2 3p\sigma^2 3p\pi^4)\Sigma^+$$

ground state may be coupled to the four first excited states (charge exchange channel)

$$(\ldots 3p\sigma^2 3p\pi^3 3d\sigma)a\ ^3\Pi,\ A\ ^1\Pi$$

and

$$(\ldots 3p\sigma 3p\pi^4 3d\sigma)b\ ^3\Sigma^+,\ B\ ^1\Sigma^+$$

either by radial, rotational, or spin-orbit couplings. In a second step, excitation takes place via couplings with higher-excited states which correlate with H(2p). A possible candidate is $3d\sigma$-$3d\pi$ rotational coupling with the configurations $(\ldots 3p\sigma^2 3p\pi^3 3d\pi)$ and $(\ldots 3p\sigma 3p\pi^4 3d\pi)$. This should result in π-state excitation only. Defining[5]

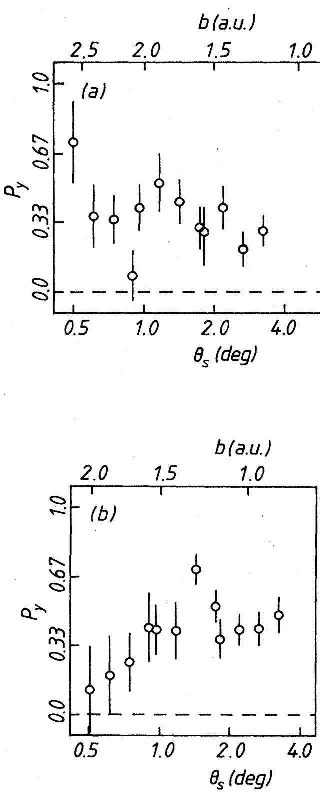

FIG. 6. Total polarization P perpendicular to the scattering plane vs scattering angle θ_s for (a) 1.5-keV and (b) 3-keV H^+-Ar collisions.

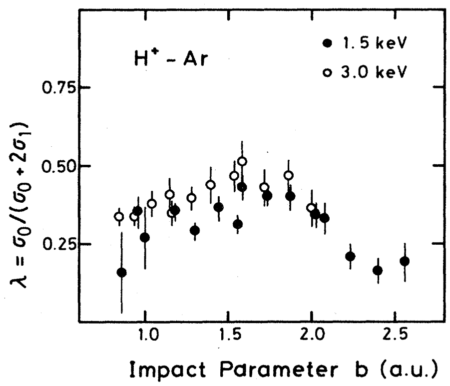

FIG. 7. Relative population of $H(2p_0)$ vs impact parameter b for 1.5-keV (●) and 3-keV (○) H^+-Ar collisions.

$$\lambda=\sum_{m_2} |f(0,m_2)|^2 \Big/ \sum_{m_2} [\,|f(0,m_2)|^2+2\,|f(1,m_2)|^2] =\sigma_0/\sigma(2p) \quad (8)$$

the relative population of the $H(2p_0)$ substate, one should expect $\lambda=0$ if it were the only excitation mechanism. Using Eqs. (2) we have $\lambda=[1-\sqrt{2}\langle T(1)_{20}\rangle/\langle T(1)_{00}\rangle]/3$. The experimental observation of $\lambda>0$ (Fig. 7) suggests that, in addition, excitation of σ states also takes place. This might be the result of a $3d\sigma$-$4p\sigma$ radial coupling, which will preferentially induce transitions near internuclear separations where the corresponding potential energy curves are closest. The calculations of Sidis[16] indicate that this internuclear separation is located at about 2 a.u. Our experimental data for 1.5-keV projectiles agree well with this prediction, as they show an increase of the relative $H(2p_0)$ population for impact parameters $b \leq 2$ a.u.

The $\langle T(1)_{22}\rangle$ alignment is connected in an interesting way to the reflection symmetry with respect to the scattering plane. From Eq. (3) we have

$$f(1,\pm 1)=+f(-1,\mp 1)\,, \qquad f(1,0)=-f(-1,0)\,. \quad (3')$$

$m_2=0$ or ± 1 leads to so-called negative or positive reflection symmetry, respectively. The sign of $\langle T(1)_{22}\rangle/\langle T(1)_{00}\rangle$ is therefore connected to the relative population of the magnetic substates of the remaining Ar^+ ion. Using Eqs. (2) and (3) one finds

$$\langle T(1)_{22}\rangle=-\,|f(1,0)|^2+2\,\mathrm{Re}[f(1,1)f(1,-1)^*]\,.$$

From this relation one immediately concludes that in the case of 1.5-keV proton impact, and at certain impact parameters where $\langle T(1)_{22}\rangle/\langle T(1)_{00}\rangle$ is positive, $H(2p_1)$ excitation goes along with preferential population of $m_2=\pm 1$ substates.

The small total polarization of about 35% results from the fact that in the present experiment the final substates of the remaining $Ar^+(^2P)$ ion cannot be specified. A total polarization of 100% may be expected only for such a collision system where the final substate of the second collision partner is also known as, for instance, in $H^+ + He \rightarrow H(2p) + He^+(1s)$ collisions. First results, in fact, show that for this collision system the total polarization is considerably higher compared to the experiments with the argon target.

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