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"Left-right asymmetry" in H(2p) charge capture from laser-oriented Na(3p)

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A strong asymmetry has been observed in 1-keV $H^+ + Na(3p) \rightarrow H(2p) + Na^+$ collisions after Na(3p) preparation with left- and right-handed circularly polarized laser light. The H(2p) state is characterized by Lyman-α photon detection. This angle-differential ion-atom-collision study shows that direct information on higher final-state multipoles can be extracted from a well-defined nonisotropic system.

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Electron exchange in kilo-electron-volt $H^+ + Na$ interactions is currently enjoying considerable interest [1–12]. Several aspects of the collision dynamics have been studied: total [1–6] and angle-differential cross sections [7–9], alignment effects [8,10,11], and the so-called "left-right asymmetry" [8,12], which is related to collisionally accumulated phases of the involved electron states [13]. This system, also being of some technical relevance, provides a convenient model case for the study of electron dynamics in ion-atom interactions; similarly to other few-electron systems [14] it may even provide a ground for the most severe ("quantum-mechanically complete") tests of our understanding of the collision. Comparatively simple situations are those in which the initial or final electron states are isotropic, e.g., in *s-p* or *p-s* transitions. For such situations, mainly two complementary experimental techniques allow "complete" studies: a nonisotropic initial state can be controlled completely, e.g., by optical pumping, and the final state is then identified by scattered-particle energy spectroscopy or by using a resonantly preferred channel [15]; in its time-inverse version, the initial state is isotropic and the nonistropic final state is characterized by its photon emission in a photon–scattered-particle coincidence [14]. A much more demanding situation is encountered if both the initial as well as the final state are nonisotropic (as, e.g., in an open-shell *p-p* transition): unless averaging over initial or final states is accepted, well-defined photon excitation of the initial state in conjunction with a photon–scattered-particle coincidence to fully characterize the final state is then required for a "complete" description.

In the present work we perform a photon-scattered particle coincidence study of the reaction

$$H^+ + Na(3p) \rightarrow H(2p) + Na^+ \quad (1)$$

at 1-keV impact energy involving angle-resolved detection of the Lyman-α fluorescence radiation from the final H(2p) state; the initial Na(3p) state is controlled by laser optical pumping. This experiment involves a fully characterized nonisotropic initial state; it is able to yield information that cannot be obtained in any other way. In this paper, we will concentrate on the "left-right asymmetry" of the capture process (Fig. 1); it has recently also been studied for the $H^+ + Na(3p) \rightarrow H(n=2) + Na^+$ reaction at 1-keV impact energy by using an energy-loss technique to identify the final $H(n=2)$ state [8,12].

The experiment will be sketched here only briefly. The basic setup has been described already earlier [5,9]; further details will be given in a forthcoming publication [16]. A 1-keV H^+-ion beam, collimated to better than 0.03° and cleaned from neutral atoms by an electrostatic deflection system, is crossed with a Na beam under single-collision conditions in a region that is free of static fields. Circularly polarized cw laser light, tuned to the $Na(3s)\,^2S_{1/2}(\bar{F}=2) \rightarrow Na(3p)\,^2P_{3/2}(F=3)$ transition, is injected into the interaction zone perpendicularly to the ion-atom plane. Lyman-α radiation (121.6 nm) emitted from capture-produced H(2p) state is detected in a solar-blind multiplier. It is placed on the ion-atom plane in the forward direction at approximately the "magic angle" ($\vartheta_\gamma = 54.7°$) relative to the ion-beam axis; viewing a length of ion beam around the interaction zone of about 10 mm, it subtends a solid angle of 0.3 sr. H particles, which are scattered through defined angles ϑ, are detected in a position-sensitive detector; it consists of a tandem multichannel plate with a divided-anode array that is composed of 16 individual segments. They cover four azimuthal angles $\varphi = 0, \pi/2, \pi, 3\pi/2 (\pm 30°)$ at each of four scattering angles ϑ; different distances between the collision zone and the particle detector allow us to span an angular rangle ϑ between 0.05° and 0.3°. Lyman-α-photon–scattered-particle coincidences select the H (2p) capture channel; they are processed in the usual way by means of 16 time-to-digital converters and a LSI 11/73

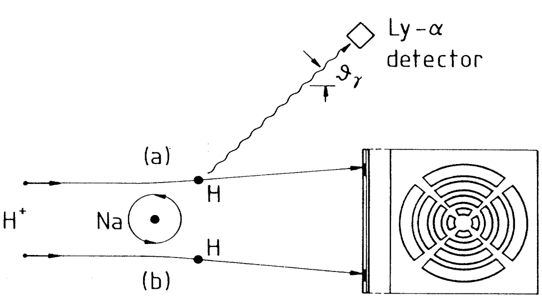

FIG. 1. Schematic representation of the interaction process and the experimental setup.

computer which also controls the experiment. Laser and Na beams are mechanically chopped; data are taken by repeatedly alternating laser-on and laser-off, as well as Na beam-on and beam-off conditions, to allow for background subtraction as well as separation of capture events from Na(3*s*) and (3*p*) states. Similarly, the circular polarization (+, −) of the laser light was repeatedly inverted; thereby, coincidence rates $I^+(\vartheta,\varphi)$ and $I^-(\vartheta,\varphi)$ were determined.

The asymmetry, defined as $A_c=(I^+-I^-)/(I^++I^-)$, is shown in Fig. 2(a) for various ϑ,φ combinations and 1-keV impact energy; the index *c* indicates that this asymmetry is the result of a particle-photon coincidence measurement. We note the fairly large values of A_c at $\varphi=0$ (closed circles) and π (open circles), particularly at small scattering angles ϑ. As a check for our experiment, we have also determined A_c at $\varphi=\pi/2$ and $3\pi/2$ (closed and open squares, respectively). In these cases, A_c must vanish due to symmetry reasons.

A full discussion of this type of experiment and its potential would require a "theory of the particle-photon coincidence experiment" for nonisotropic initial states,

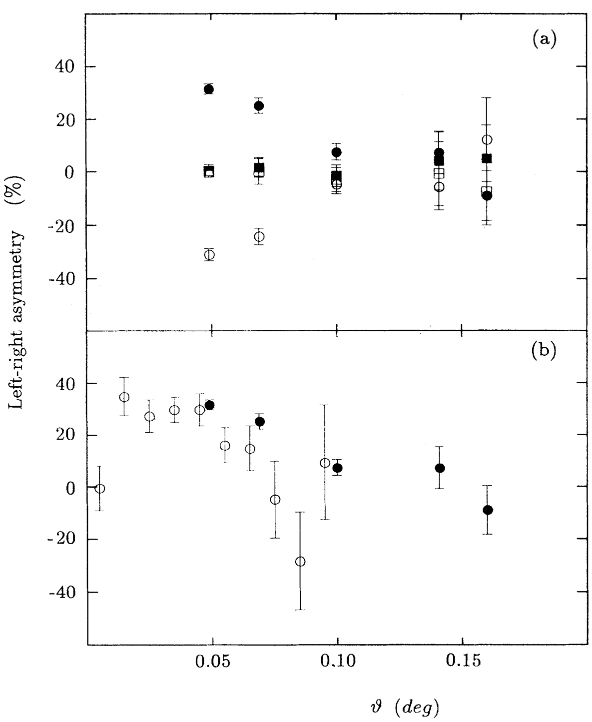

FIG. 2. The left-right asymmetry in 1-keV $H^++Na(3p)$ collisions vs scattering angle ϑ. (a) Asymmetry $A_c=(I^+-I^-)/(I^++I^-)$ for H(2*p*) capture as obtained from a particle-photon coincidence experiment at azimuthal scattering angles $\varphi=0$ (●), $\pi/2$ (■), π (○), $3\pi/2$ (□); I^+ (I^-) are the coincidence rates for left (right) circularly polarized pumping light preparing the initial Na(3*p*) state. (b) Asymmetry $\overline{A}_c$ (●); A (○) are data from Houver *et al.* [12] for H($n=2$) capture. $\overline{A}_c$ may be understood as the average of the absolute asymmetries obtained at $\varphi=0$ and π in a particle-photon coincidence experiment involving a H(2*p*) final state; A is the asymmetry as obtained in a noncoincident experiment with a H($n=2$) final state (for the exact definitions of $\overline{A}_c$ and A, see text).

similarly to the one developed, for example, by Blum and Kleinpoppen (cf., e.g., Ref. [17]), which is generally used for isotropic initial states. This will be the subject of a more extensive forthcoming paper [16]. The present discussion of the left-right asymmetry is greatly facilitated since the scattering plane coincides with the symmetry plane of the initial Na(3*p*) state. Thus the collision situations (+ circular polarization, $\varphi=0$) and, correspondingly, $(-,\pi)$ are physically equivalent; in both cases, projectile and active electron move *antiparallel* around the Na core (cf., e.g., case *a* in Fig. 1). Similarly, the combinations $(-,0)$ and $(+,\pi)$ would correspond to a case *b*, in which projectile and electron move *parallel.* An experiment in which the final state is identified by an energy-loss measurement (as performed, e.g., by Houver *et al.* [12]) gives direct information on the differential scattering cross section, i.e., the lowest-order final-state multipole $\langle T_{00}^+\rangle$ (for the definition of state multipoles, cf., e.g., Ref. [17]); it will thus produce symmetric results for $\varphi=0$ and π: $A(\varphi=0)=-A(\varphi=\pi)$, with A defined correspondingly. In contrast, the photon-particle coincidence requirement applied in the present work introduces an asymmetry: The photon detector views the final H(2*p*) state from a well-defined direction and thus collects the corresponding additional information on its charge, rotation, and location in space. As a consequence, for a fixed photon observation angle φ_γ, $A_c(\varphi=0)\neq -A_c(\varphi=\pi)$, in general [18]; only if *all* Lyman-α photons were to be collected (4π solid angle) would both techniques produce the same results, $A_c\equiv A$. The left-right asymmetry measurement can be analyzed further without elaborate calculations simply by applying Eq. (50) from Blum and Kleinpoppen [17]; this relation can be interpreted as the coincident Lyman-α emission rate for decay of a H(2*p*) state that is assumed to be space fixed and viewed by a photon detector under various angles. It thereby is assumed that the processes of Na optical excitation, charge capture, and H(2*p*) deexcitation can be treated separately. The H(2*p*)-state multipoles $\langle T_{KQ}^+\rangle$ are combinations of the various amplitudes $f_{\mu\nu}$ linking the initial Na($3p_\mu$) and final H($2p_\nu$) states ($\mu,\nu=0,\pm1$); besides the scattering angle, they depend in general also on the azimuthal scattering angle φ due to the nonspherical symmetry of the initial Na(3*p*) states [16]. The $\langle T_{20}^+\rangle$ term is eliminated from Eq. (50) by $\vartheta_\gamma\approx 55°$; abbreviating $\langle T_{KQ}^+\rangle$ by T_{KQ}, Eq. (50) reduces (for a given scattering angle φ) to

$$I=c_0T_{00}-c_1T_{22}\cos 2\varphi_\gamma+c_2T_{21}\cos\varphi_\gamma\ , \tag{2}$$

where c_0, c_1, c_2 are constants containing Clebsch-Gordan coefficients, and φ_γ is the azimuthal photon-detection angle. Due to the symmetry indicated above, measurements at $\varphi=0,\pi$ and fixed $\varphi_\gamma=\pi$ can be viewed as measurements at fixed $\varphi=0$ and $\varphi_\gamma=\pi,0$. Redefining $A_c=(I^a-I^b)/(I^a+I^b)$ to eliminate the sign inversion when going from $\varphi=0$ to π [with *a* (*b*) referring to the cases antiparallel (parallel) motion as defined above], we obtain

$$A_c(\varphi=0)=\frac{c_0\delta_{00}-c_1\delta_{22}+c_2\delta_{21}}{c_0\sigma_{00}-c_1\sigma_{22}+c_2\sigma_{21}}\ , \tag{3a}$$

$$A_c(\varphi=\pi)=\frac{c_0\delta_{00}-c_1\delta_{22}-c_2\delta_{21}}{c_0\sigma_{00}-c_1\sigma_{22}-c_2\sigma_{21}} , \quad (3b)$$

where $\delta_{KQ}=T^a_{KQ}-T^b_{KQ}$ and $\sigma_{KQ}=T^a_{KQ}+T^b_{KQ}$. As already stated, $A_c(0)\neq A_c(\pi)$.

For the purpose of a further brief discussion we will assume small H(2p) anisotropy. Then $\Delta=A_c(0)-A_c(\pi)\approx 2c_2\delta_{21}/c_0\sigma_{00}$; within our experimental uncertainty, $\Delta\sim 0$ at most angles ϑ investigated (except possibly at 0.1°). We mention that $tg\gamma=2T_{21}/(T_{22}-\sqrt{2}T_{20})$, with γ the H(2p) "alignment angle" [19]; small Δ could thus be an indication that $\gamma^a\approx\gamma^b$, although a more conclusive discussion would also require knowledge about the behavior of the other T_{KQ}, i.e., measurements at other photon observation angles. Furthermore, $\overline{A}_c=[A_c(0)+A_c(\pi)]/2\approx\delta_{00}/\sigma_{00}=A$, i.e., the result expected from an energy-spectroscopy experiment. We have plotted $\overline{A}_c$ in Fig. 2(b) and compare it to the A values obtained by Houver *et al.* [12], although it has to be kept in mind that their experiment selects the entire H(n =2) state. In the region of overlap, both data sets behave similarly; this may not be too surprising since calculations by Shingal [20] indicate dominance of H(2p) over the H(2s) channel up to $\vartheta\approx 0.075°$. Note that the sign of $\overline{A}_c$ confirms the finding of Ref. [12] of a behavior opposite to a simple "rolling-ball" picture unless negative (attractive) projectile deflections were invoked at the very small ϑ values here. Such a result would not be unexpected since the asymmetry depends on collisionally accumulated phases that may show a quite complex behavior at the low impact energy of 1 keV; in this quasimolecular regime, deviations from the rolling-ball model are also well known from other coherence studies in ion-atom collisions (cf., e.g., [14]).

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