Strong Right-Left Asymmetry Observed in Charge Transfer from Circular Atomic States near the Matching Velocity

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A strong right-left scattering asymmetry, depending on scattering angle, has been observed for electron capture by protons impinging on circular states of Na($3p$), excited by circularly polarized light incident perpendicular to the scattering plane. Scattering angles are extremely small, corresponding to grazing incidence collisions. Proton energy is 1 keV, i.e., a collision velocity about half of the orbital velocity of the sodium $3p$ electron. The observed asymmetry agrees with recent elaborate theoretical predictions using large atomic basis sets.

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The role of orientation in atomic collision dynamics, illustrated classically by the sense of rotation of the active electron around the atomic core, has been discussed intensively in recent years [1–3].

The use of left and right circularly polarized laser light for preparation of oriented, excited atomic states (maximum electron angular momentum projection) prior to the collision has initiated a series of crossed beam experiments [4–6]. The resulting right-left asymmetry of the differential scattering cross sections (see Fig. 1) was first studied for heavy particle collisions at thermal energies by Hertel and co-workers [4] for rovibrational energy transfer in Na($3^2P_{3/2}$)-N_2 collisions, and by Düren and co-workers [5] in the scattering of excited K($4^2P_{3/2}$) atoms from argon. These data highlight the role of coherence in the scattering process and the sensitivity to the long-range part of the potential. A pronounced right-left asymmetry was observed for 10–100-eV Na^+ ions scattered from oriented Na($3^2P_{3/2}$) atoms by Schmidt *et al.* [6] in direct excitation and deexcitation reactions leading to Na($3d$) and Na($3s$), respectively. This observation was interpreted in terms of the phase difference of the amplitudes accumulated along the quasimolecular Σ and Π potentials at large internuclear distances [7,8]. Small but significant orientation effects varying with scattering angle in a resonant charge-transfer process were reported the first time for 50–100-eV energy Na^+-Na($3^2P_{3/2}$) collisions by Witte *et al.* [9].

At larger energies, defined here as the region where the collision velocity v_c becomes comparable to the velocity v_e of the active electron, orientation in heavy particle collisions was first investigated in the time-reversed scheme in experiments with planar scattering symmetry where the electronic state of a collisionally excited atom is investigated by polarization analysis of the photons emitted *after* the collision [10–12]. Propensity rules for orientation, i.e., a strongly preferred sense of circulation of the active electron around the atomic core, were formulated for direct excitation by Andersen and Nielsen [11] and confirmed in several experiments [12]. The possibility to extend such a description to charge-transfer processes is a delicate question, since the active electron now changes center during the collision and the treatment should therefore include the corresponding momentum transfer to the electron. A classical picture modeling the electron-capture process in terms of simple velocity matching, suggesting that electron transfer is favored when the projectile and the active electron move in the same direction on the same side of the nucleus, was invoked for the description of charge transfer for protons colliding with state-selected hydrogenic Rydberg atoms [13] at $v_c \geq v_e$. Very recently Roncin *et al.* [14] have observed a strong propensity for orientation of $B^{2+}(2p)$ atoms produced by electron capture in $B^{3+}(1s^2)$-He collisions at velocities $v_c \approx 0.1 v_e$, which follows the velocity matching picture and the theoretical prediction for this system by Hansen *et al.* [15].

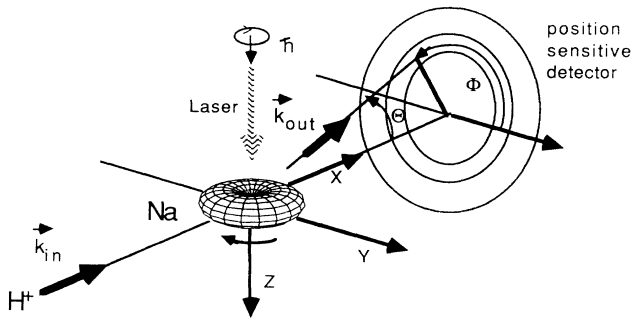

FIG. 1. A schematic diagram of the experiment for investigation of right-left scattering asymmetry on oriented atoms. A beam of (left-hand or right-hand) circularly polarized photons excite and orient Na atoms as shown. Protons incident along the x axis may capture the excited electron and are scattered in the direction $\mathbf{k}_{out}$. The neutral H atoms are detected by a position-sensitive device.

Experimental developments for the H^+-Na($3p$) system [16–18] and quantitative predictions of orientation effects in the $v_c \approx v_e$ velocity range have been performed recently. Differential cross sections predicting an orientation effect for charge transfer to the H($n=2$) channel have been obtained by Courbin and co-workers [19] in a nineteen-state molecular treatment including electron translation factors. A general study of orientation propensity rules in charge-transfer processes with full inclusion of electron translational factors has been done by Nielsen and co-workers [20] in a two-center atomic basis and applied to the H^+-Na($3p$) system in a ten-state calculation.

This paper reports to our knowledge the first experimental study of effects of initial-state orientation in a charge-transfer process in the velocity matching region. For the collision energy $E=1$ keV ($v_c=0.4v_e$) we have observed a large orientation effect for the nonresonant charge-exchange reaction ($\Delta E=0.36$ eV)

$$H^+ + Na(3p, m_l = \pm 1) \rightarrow H(n=2) + Na^+ , \qquad (1)$$

revealed as a strong right-left asymmetry of the differential scattering cross sections, in the sense *opposite* to what is expected from the naive velocity matching picture but in agreement with recent theoretical predictions based on atomic calculations [20,21].

For the H^+-Na($3p$) quasi-one-electron system we earlier observed a strong dependence on the initial $3p$ orbital alignment in total cross sections for electron capture into the H($n=2$) and H($n=3$) states [18]. The experimental setup used was described in detail in Ref. [16]. The present investigation of orientation effects has required a series of refinements on the experimental side, in particular since the neutral H atoms are forwardly scattered in an extremely narrow angular cone, $\Delta\theta < 0.2°$: (i) The angular aperture of the incident ion beam of energy E is reduced to $\Delta\theta_i \approx 0.04°$ (FWHM), and (ii) the scattered neutrals are now detected by a position-sensitive device [22] which enables accurate determination of double-differential cross sections $\sigma(\theta,\phi)$, where θ is the scattering angle and ϕ the azimuthal angle which defines the collision plane (see Fig. 1). The detector consists of three channel plates and a square resistive anode and is located at a distance of 4.50 m from the collision region, which corresponds to an angular acceptance of 0.5°. The sodium beam is produced in a quasisupersonic expansion. The laser light is circularly polarized, alternatively right- and left-handed (negative and positive photon helicity, respectively, following the convention of classical optics [2]) by means of a Pockels cell inserted in the light path, and locked to the [$3^2S_{1/2}(F=2) \rightarrow 3^2P_{3/2}(F=3)$] hyperfine transition. This pumping scheme leads to steady-state population of $m_F=-3$ ($m_L=-1$) and $m_F=+3$ ($m_L=+1$) states, respectively. For each detected event, we record three coordinates: the time-of-flight (T), the position (x,y) of the scattered particle, and in addition an index i characterizing the collision (laser on/off, polarization right/left, sodium beam on/off).

Details of the data analysis will be reported in a forthcoming publication. Only the main steps are given here. A first selection of the events by the index i provides histograms for the number of counts I as a function of position for the H^+-Na($3s$) collision, and for the H^+-Na($3p$) collision after the weighted subtractions $I^+_{on} - (1-\alpha)I_{off}$ and $I^-_{on} - (1-\alpha)I_{off}$, where I_{off}, I^+_{on}, and I^-_{on} are the number of neutral H atoms scattered without (off) and with (on) laser for left- (+) and right- (−) hand circular polarization of the laser, respectively. The effective fraction α of excited Na($3p$) atoms [23] is typically 10%. In order to interpret the scattering process in terms of doubly differential cross sections $\sigma(\theta,\phi)$, the symmetry center of the distributions on the detector has to be accurately determined. This is done with reference to the I_{off} histogram since the target is isotropic in this case. A second selection of the events is provided using the time of flight information, since the dominant reactions taking place, (1) and ($\Delta E=-1.75$ eV)

$$H^+ + Na(3s) \rightarrow H(n=2) + Na^+ , \qquad (2)$$

are discriminated by the time or energy resolution of the spectrometer, $\Delta E \approx 1$ eV at $E=1$ keV. Because of this energy resolution the histogram which corresponds to reaction (1) will not depend on the fraction of excited states α.

The $\sigma^{\pm}(\theta,\phi)$ differential cross sections can be parametrized in the following way:

$$\sigma^{\pm}(\theta,\phi) = C_0(\theta) \pm C_1(\theta)\cos\phi + C_2(\theta)\cos 2\phi . \qquad (3)$$

The parameters $C_i(\theta)$ are known functions of the scattering amplitudes $f_{\lambda\Sigma}(\theta)$, $f_{\lambda\Pi^+}(\theta)$, and $f_{\lambda\Pi^-}(\theta)$ for the collision process where, in a molecular representation, the initial state is Σ, Π^+, or Π^- and the final state λ is one of the $2s\Sigma$, $2p\Sigma$, $2p\Pi^+$, and $2p\Pi^-$ final states of the H($n=2$) + Na^+ channel:

$$C_0(\theta) = \tfrac{1}{2}\{\sigma_\Sigma(\theta) + \tfrac{1}{2}[\sigma_{\Pi^+}(\theta) + \sigma_{\Pi^-}(\theta)]\} ,$$

$$C_1(\theta) = \mathrm{Im}\left[\sum_\lambda f_{\lambda\Sigma}(\theta) f^*_{\lambda\Pi^+}(\theta)\right] ,$$

$$C_2(\theta) = \tfrac{1}{4}[\sigma_{\Pi^+}(\theta) - \sigma_{\Pi^-}(\theta)] ,$$

$\sigma_\Sigma(\theta)$, $\sigma_{\Pi^+}(\theta)$, and $\sigma_{\Pi^-}(\theta)$ are the differential cross sections for the production of H($n=2$), where $\sigma_\Sigma(\theta) = \sum_\lambda |f_{\lambda\Sigma}(\theta)|^2$, etc.

Figure 2 displays differential cross sections obtained for the left- and right-hand circular polarization of the laser light, integrated over azimuthal angle ϕ in four sectors of 90° as indicated. This figure clearly demonstrates strong preferential electron capture at this energy for a proton traveling in the *opposite* direction to the orbiting electron as indicated in the upper panel, assuming a repulsive interaction.

For further quantitative analysis of the anisotropy effects, we Fourier analyze the data as a function of the

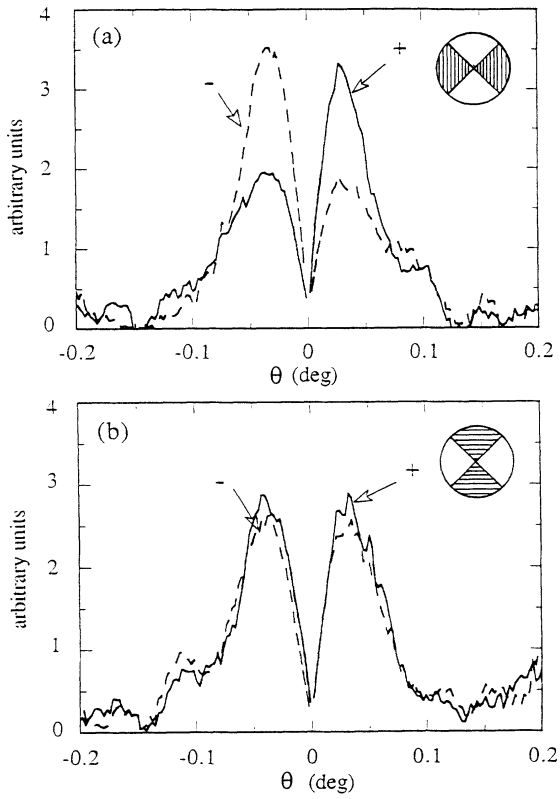

FIG. 2. The number of hydrogen atoms detected in the four sectors indicated, as a function of the scattering angle for left-hand circular (+, ——) and right-hand circular (−, ---) polarized photons, respectively. (a) The strong right-left asymmetry. (b) The expected up-down symmetry.

azimuthal angle; cf. Eq. (3). From this fit we derive the right-left asymmetry parameter $A(\theta)$, Fig. 3:

$$A(\theta)\equiv\frac{I^+(\theta,0)-I^+(\theta,\pi)}{I^+(\theta,0)+I^+(\theta,\pi)}=\frac{I^+(\theta,0)-I^-(\theta,0)}{I^+(\theta,0)+I^-(\theta,0)}$$
$$=\frac{C_1(\theta)}{C_0(\theta)+C_2(\theta)}. \quad (4)$$

$A(\theta)$ is positive at the smaller angles, reaches a maximum value of 0.35, and then shows a dip to negative values at $\theta\approx 0.085°$. A similar behavior was found for $E=0.75$ and 1.5 keV. The asymmetry parameters deduced from molecular [19] and recent atomic [20] calculations are also reported in Fig. 3. The results of the atomic calculation reproduce the experimental trends nicely while the molecular calculation differs considerably. A detailed analysis of the underlying physics of the observed behavior and its apparent variance with the velocity matching picture goes beyond the space limit imposed by this Letter but will be discussed in connection with the presentation of detailed differential cross sections [21].

In conclusion, we have measured a strong right-left scattering asymmetry for electron capture for protons colliding with Na($3p$) atoms prepared in circular states. The present investigation of scattering asymmetries has required a series of experimental refinements, in particular since the neutral H atoms are forwardly scattered in an extremely narrow angular cone. Future experiments will concentrate on expansion of the velocity range and determination of a complete set of parameters describing also the shape and the alignment angle of the charge cloud of the active electron. This will further advance the understanding of charge-transfer dynamics and the role of electron-charge-cloud orientation in the difficult energy range where the collision velocity and the electron velocity are of similar size.

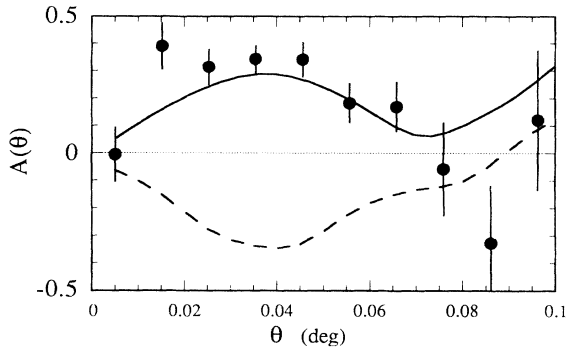

FIG. 3. The asymmetry parameter defined in Eq. (4) as a function of scattering angle: (——) nineteen-state AO theory; (---) nineteen-state MO theory; (●) present experiment. Error bars show statistical uncertainty from the fitting procedure (twice the standard deviation). The two theoretical curves have been convoluted with the experimental resolution.

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