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Left-right scattering asymmetries for electron transfer from oriented and tilted aligned Na$(3p)$ states to H$(n=2,3)$

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We present experimental and theoretical results for the spatial pattern of forwardly scattered H$(n=2,3)$ atoms created by electron transfer from optically prepared Na$(3p)$ atoms by 2-keV proton impact. The near-resonant H$(n=2)$ and the endoergic H$(n=3)$ channels display dramatically different left-right scattering asymmetries, varying strongly with the initial polarization of the target. The results illuminate current ideas on the role of the internal dynamics for transition propensities in the electron transfer process, including a connection between the collisional behavior of aligned and oriented states. [S1050-2947(96)05207-9]

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The control of the shape and dynamics of optically prepared electronic states has enabled a number of studies of charge transfer at a very fundamental level [1]. Experimental and theoretical results have recently been obtained in the ''velocity-matching region'' where the collision velocity, v_c, and the velocity of the active electron, v_e, are comparable.

In this context we present results for electron transfer from oriented and tilted aligned Na$(3p)$ states into H$(n=2,3)$ in H$^+$-Na collisions. These results enable a direct comparison of a slightly exoergic and an endoergic electron transfer reaction, the population of H$(n=2)$ and H$(n=3)$, respectively. Strongly contrasting left-right asymmetries are observed in the plane of scattering from an oriented p state, as well as from a ''dumbbell''-shaped p orbital tilted by an angle with respect to the incoming proton beam, and are interpreted using coupled channel calculations. The role of orbital orientation, i.e., the sense of circulation of the active electron around the atomic core, is discussed in terms of general models, particularly the velocity-matching argument of Kohring *et al.* [2] and the propensity rule for orientation explored by Nielsen *et al.* [3]. The relationship between the alignment and orientation effects, as recently proposed by Machholm and co-workers [4,5], is discussed.

Two types of observables are relevant for the investigation of the role of alignment and orientation in electron transfer from prepared electronic states: total cross section (TCS) and angular differential cross section (DCS) probing, e.g., left-right scattering asymmetries.

Total electron transfer cross sections for Na$^+$ ions impinging on circular and elliptic Li Rydberg atoms have been measured at velocities $v_c > v_e$ [6] and found to be consistent with the velocity-matching argument [2]. They are also in good agreement with predictions of recent classical trajectory Monte Carlo (CTMC) [7] and coupled channel calculations [8]. Alignment effects on TCS have been observed in an extended velocity range for electron capture by singly [9] and multiply charged ions [10–12] from a Na$(3p)$ orbital aligned parallel or perpendicular to the collision velocity. The experimental trends are well reproduced by semiclassical coupled channel and CTMC calculations [9–15]. They show that for velocities above the matching velocity [$v_e=0.47$ a.u. for Na$(3p)$ from the virial theorem], electron transfer is strongly favored for a p orbital initially aligned parallel to the collision velocity. A simple model based on the overlap of the relevant target and projectile wave functions in momentum space [14] reproduced this dependence well for He^{2+} [10] (though not for O^{6+} [11]). This supports an interpretation of the orbital alignment effect as a kinematical effect at velocities above v_e.

Previously, angular resolved scattering patterns for quasiresonant, state-selected electron transfer in collisions between H$^+$ [16,17], He$^+$ [18], Li$^+$ [18], and H$_2$$^+$ [19], and an optically prepared Na$(3p)$ target have been measured. The DCS for velocities smaller than v_e revealed strong spatial anisotropies. A complete density-matrix determination for each reaction channel as a function of the scattering angle θ enables a very sensitive test of theory [20].

We report an angular resolved experimental and theoretical study of the reactions

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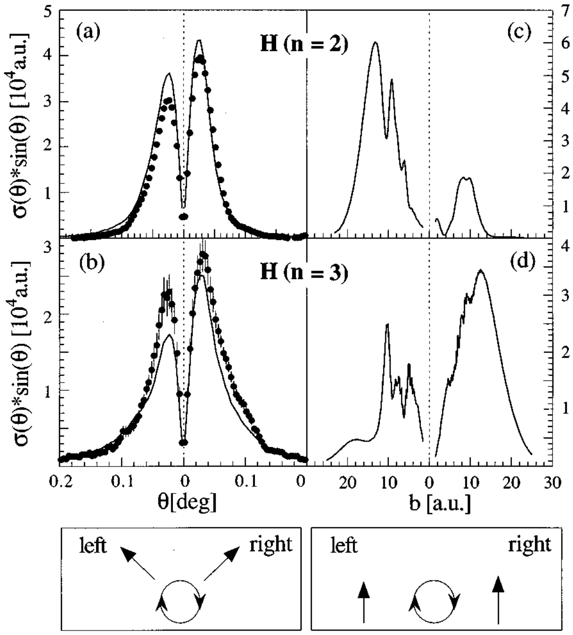

FIG. 1. Reduced differential cross sections $\sigma(\theta)\sin\theta$ versus scattering angle θ (a), (b) and semiclassical probabilities $P(b)$ versus impact parameter b (c), (d) for electron capture into H($n=2$) (a), (c) and H($n=3$) (b), (d) from a Na($3p_{-1}$) state for left-right scattering and for left-right passage of the projectile, respectively, as shown schematically. In (a) and (b), experimental data (dots) and theoretical data (line) are compared, as discussed in the text.

$$\mathrm{H}^+ + \mathrm{Na}(3p) \rightarrow \mathrm{H}(n=2) + \mathrm{Na}^+, \quad \Delta E = -0.36 \text{ eV} \quad (1)$$

$$\mathrm{H}^+ + \mathrm{Na}(3p) \rightarrow \mathrm{H}(n=3) + \mathrm{Na}^+, \quad \Delta E = +1.52 \text{ eV} \quad (2)$$

at the impact energy $E=2$ keV or $v_c=0.28$ a.u. ΔE is defined as positive for an endoergic reaction. By means of a circularly or linearly polarized laser beam incident perpendicular to the ion beam, a Na($3p$) state is prepared as an oriented state, Na($3p_{\pm 1}$), labeled by the angular momentum projection $m_l = \pm 1$ according to the sense of rotation of the valence electron around the atomic core, or an aligned p orbital tilted by an angle $\gamma = 45°$ with respect to the beam direction, in the following denoted by Na($3p_{45}$). At the collision energy chosen, the two reactions are of comparable magnitude, and they exhibit opposite TCS alignment effects [21]: electron transfer to H($n=2$) is favored for an orbital initially aligned parallel to the proton velocity, and to H ($n=3$) for an orbital aligned perpendicular to the proton velocity. For reaction (1) differential cross sections measured for scattering from an oriented state [16] compared well with coupled-channel calculations using a basis of atomic [22] or molecular orbitals [5]. Orientation effects were found to be consistent with the predictions of the velocity-matching concept. Polarization effects for this system have also been discussed in the frame of the asymptotic theory for charge transfer [23].

In the experiment an ion beam, a Na atom beam, and a laser beam with an electro-optical modulator cross each other at right angles; for details, see [16] and [24]. Time-of-flight spectroscopy enables separation of channels (1) and (2). Neutral atoms are scattered forwardly in a very narrow cone, $\theta < 0.2°$, and are detected with a position-sensitive device. The density-matrix elements are extracted from a Fourier analysis of the scattering pattern $N(\theta,\phi)$, where the azimuthal angle ϕ identifies the collision plane for each event detected [16]. The Na($3p_{-1}$) and Na($3p_{45}$) initial orbital geometries chosen here provide complementary information on the scattering process, respectively, the imaginary and the real parts of the coherence terms of the density matrix [16].

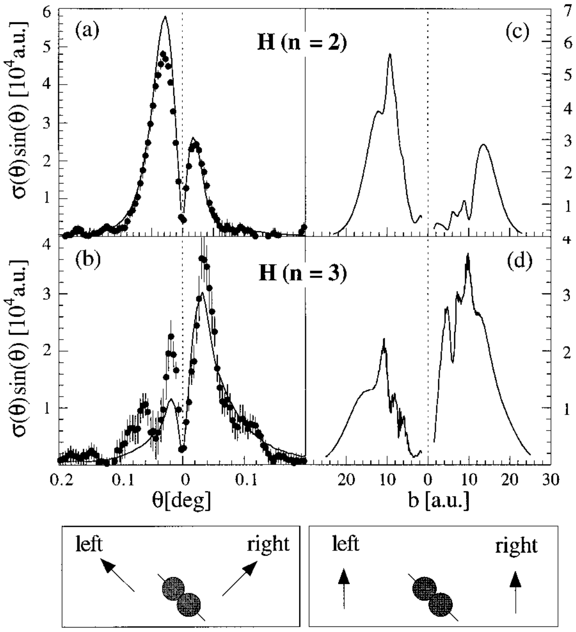

FIG. 2. Reduced differential cross section $\sigma(\theta)\sin\theta$ versus scattering angle θ (a), (b) and semiclassical probabilities $P(b)$ versus impact parameter b (c), (d) for electron capture into H($n=2$) (a), (c) and H($n=3$) (b), (d) from an aligned Na($3p_{45}$) orbital, for left-right scattering and for left-right passage of the projectile, respectively, as shown schematically. In (a) and (b), experimental data (dots) and theoretical data (line) are compared, as discussed in the text.

The theoretical results are based on the coupled-channel impact parameter method using a straight-line projectile trajectory. The electronic wave function is expanded on a 36-state atomic basis including electron translational factors and describes adequately the reaction channels under consideration. The differential cross sections are obtained from the semiclassical probability amplitudes by an eikonal transformation [22,25].

We present the data in the form of reduced differential cross sections, $\sigma(\theta)\sin\theta$, for H($n=2$) and H($n=3$) scattering in the orbital plane of an oriented Na($3p_{-1}$) state (Fig. 1) and a Na($3p_{45}$) aligned orbital (Fig. 2). The experimental results are compared to the theoretical results convoluted with the apparatus function, and are normalized using the calculated TCS for electron capture from a Na($3s$) target, taking into account the effective fraction of excited states, $\alpha \simeq 0.40$. The angular collimation of the ion beam is $\Delta\theta = 0.04°$ full width at half maximum. The corresponding semiclassical probabilities as a function of impact parameter b are shown for comparison.

Figures 1(a) and 1(b) display the reduced DCS for reactions (1) and (2) for scattering on a Na($3p_{-1}$) state. It shows a left-right asymmetry with a preferred scattering to the right for both processes (1) and (2). The experimental and calculated DCS agree reasonably well. It is therefore instructive to address the corresponding semiclassical probabilities for the two reactions, Figs. 1(c) and 1(d). They show a very different behavior: electron transfer to H($n=2$) is strongly favored for a left-side trajectory for which the incoming projectile ion and the target electron have velocities in the same direction, in agreement with the velocity-matching concept. Transfer to H($n=3$), however, is favored for a right-side trajectory, for which the ion velocity and the orbital velocity point in opposite directions.

Figure 2 displays similar results, but now for scattering on a Na($3p_{45}$) tilted orbital. The overall agreement between the experimental and theoretical observables is again reasonable for the dominant features, although oscillations seen in the measured DCS are not reproduced in the calculations. It shows an even more pronounced left-right asymmetry, with a preferred scattering to the left for electron transfer to H($n=2$) (a), and to the right for electron transfer to H($n=3$) (b). The corresponding semiclassical probabilities, Figs. 2(c) and 2(d), again show a very different behavior: electron transfer to H($n=2$) is strongly favored for a left-side trajectory while transfer to H($n=3$) is favored for a right-side trajectory.

We point out that when comparing Figs. 1 and 2 the strong left-right asymmetry shown in the similar looking semiclassical probabilities $P(b)$ clearly prevails in the DCS for capture from the tilted orbital (Fig. 2), but is strongly attenuated for capture from the circular state (Fig. 1). We have recently observed a similar behavior for the He$^+$,Li$^+$-Na($3p$) systems [18].

We turn to the interpretation of these results in terms of general models, using the semiclassical probabilities. Two main conclusions can be drawn:

(i) The first one addresses the role of orbital orientation. For the quasiresonant electron transfer from a Na($3p_{-1}$) state to H($n=2$), the process is favored for a left-side trajectory, in agreement with the velocity-matching concept. The state-to-state calculations predict a dominant charge transfer from the Na($3p_{-1}$) state into H($2p_{-1}$) at large impact parameters. This prediction is analogous to the $3p_{-1}\rightarrow 3p_{-1}$ preference found in Na$^+$-Na($3p$) resonant electron transfer, where the velocity-matching criterion was related to the role of the electron translational factors in the theoretical description [26]. Recent CTMC calculations with a Na($3p_{-1}$) target state reproduce qualitatively features of the left-right asymmetries observed here [27].

For the endoergic electron transfer from a Na($3p_{-1}$) state to H($n=3$), the process is, in contrast, favored for a right-side trajectory, corresponding to projectile and electron velocities having opposite directions. For a nonresonant process, one should compare with the orientation propensity criterion [3]. In its simplest form, it predicts that at velocities v_c near the maximum of the TCS for a specific electron transfer process, the following criterion is satisfied (in atomic units):

$$\left(\Delta E+\frac{v_c^2}{2}\right)\frac{a}{v_c}\pm\pi\Delta m\simeq 0, \tag{3}$$

where the ± signs refer to left- and right-side trajectories, respectively, at large impact parameters. $\Delta m=m_{final}-m_{initial}$ is the preferred change of electron orbital angular momentum projection on a quantization axis perpendicular to the collision plane, and a is the effective interaction length. At 2-keV proton impact energy, the effective energy defects ($\Delta E+v_c^2/2$) for reactions (1) and (2) are equal to 0.73 eV and 2.61 eV, respectively, i.e., the quasiresonant character of reaction (1) remains and the endoergic character of reaction (2) is enhanced. For capture into H($n=3$), where the process is favored for a right-side trajectory, criterion (3) predicts a preferred change of orientation $\Delta m>0$. The present result for electron transfer from the Na($3p_{-1}$) state is consistent with this prediction, since several final states with $m_f>-1$ are accessible. Indeed for this process, dominance of Na($3p_{-1}$)$\rightarrow$H($3p_{+1}$) and $H(3s)$ is found in the state-to-state calculations for right-side trajectories, corresponding to $\Delta m=+2,+1$, in agreement with the propensity criterion (3).

(ii) The second conclusion addresses a possible relationship between the collisional behavior of aligned and oriented states. We observe that from both a Na($3p_{-1}$) and a Na ($3p_{45}$) initial state, electron transfer to H($n=2$) is favored for a left-side trajectory and electron transfer to H($n=3$) is favored for a right-side trajectory. This behavior can be related to results obtained by Machholm and Courbin in a theoretical study of the time evolution of the valence electron density and current along the trajectory for 1-keV Li$^+$-Na($3p$) collisions [4]. In their study, a Na($3p_{45}$) orbital gradually acquires a *negative* orientation, induced by the approach of the projectile ion both for a left- and a right-side trajectory. The opposite left-right asymmetries found for reactions (1) and (2) [Figs. 2(c) and 2(d), respectively] for an initial Na($3p_{45}$) orbital may thus be correlated with the similar asymmetries found for the Na($3p_{-1}$) initial state [Figs. 1(c) and 1(d)]. This dynamical process which invokes a relationship between the alignment and orientation effect is distinct from the kinematical model for the alignment effect at velocities above v_e, formulated in terms of momentum space wave-function overlap [14].

In conclusion, left-right scattering asymmetries have been observed for the production of H($n=2$) and H($n=3$) atoms in electron transfer from a target in the Na($3p_{-1}$) or Na($3p_{45}$) state, and interpreted using semiclassical coupled-channel calculations. The analysis of the left-right scattering asymmetries reaffirms the ''velocity-matching'' concept for *quasiresonant* processes, and the orientation propensity criterion, Eq. (3), for *nonresonant* processes. The correlation noted between the alignment and orientation effects supports the interpretation of the role of orbital alignment as a dynamical effect for $v_c<v_e$. In this paper we have concentrated on the Na($3p_{-1}$) and Na($3p_{45}$) scattering geometries. Results for the complete density-matrix determination for reactions (1) and (2), including the initial-state aligned parallel and perpendicular to the ion-beam direction as well as state-to-state semiclassical results and CTMC calculations will be presented in forth-

coming papers. Experiments aiming at final-state identification in TCS and DCS measurements are also under development [28]. Further systematic studies of a broader class of collision systems over a wider range of collision velocities are desirable to explore the limits of the present models.

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