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Anomalous final-state distributions of electrons captured from directed Rydberg states

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A classical model of singly charged ion collisions with Rydberg atoms in linear Stark states directed toward the oncoming ion beam predicts a bimodal distribution of final states resulting from electron capture. At a reduced velocity $\tilde{v}=v_{\rm ion}/v_{\rm Bohr}=0.45$, the calculated energy distribution of captured electrons peaks near the target binding energy E_i, but has a second peak at energies roughly $E_i/4$. Analysis of the impact-parameter dependence of the capture probabilities indicates that the second peak arises from three-swap capture events. The decrease in binding energy of the captured electrons is attributed to excitation via united-atom rotational coupling. [S1050-2947(98)50401-5]

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Capture of electrons from Rydberg atoms by singly charged ions is an important three-body process and is an active and robust area of experimental research. Electron-capture cross sections have been measured near matching velocity from laser-aligned [1,2], circular [3], elliptic [4–7], and linear Stark [8,9] Rydberg targets. These targets offer a unique opportunity to study Coulomb three-body rearrangement in the correspondence principle regime where quantum-mechanical calculations are currently intractable.

One important recent development has been the recognition that multiple passes (or swaps) of the electron between the nuclei occur at intermediate velocities. Structure in the total capture cross section for singly charged ions striking Na atoms in 24*d* or 24*s* states has been attributed to partially resolved contributions of one-, three- and higher-odd-swap processes [1]. For reduced velocities $\tilde{v}\approx 1$, one-swap processes are dominant, owing to the ''velocity matching'' of the target electron and the projectile ion near the closest approach [10–18]. Here, three-swap processes contribute only weakly. However, for slightly lower ion speeds, $\tilde{v}\approx 0.5$, the electron has sufficient time to pass three times between the charge centers, and a prominent three-swap feature appears in the total cross sections [1]. At very high velocities, three-swap trajectories also contribute significantly, accounting for the Thomas mechanism [19], while for very low velocities, $\tilde{v}\ll 1$, the electron passes many times between the charge centers in an analog of the resonant charge-transfer processes long studied using ground-state targets [20].

Classical calculations of charge transfer are used here to point out that three-swap contributions to capture result in heretofore unnoticed structure in the distribution of final capture states. In particular, a high-energy peak in the distribution of final states is found, due to small-impact-parameter three-swap collision processes. Previous experimental studies [21–23] of the final-state energy distribution of electrons captured from Rydberg targets in low-angular-momentum eigenstates showed no evidence of such structure and none was noted in the classical calculations of Pascale *et al.* [24] An indication of a secondary peak at high *n* was apparent, however, in classical calculations by MacKellar [25], and at the time it was suspected to be a Coriolis effect [26]. Our classical analysis predicts, however, that this structure is far more apparent if the target is prepared in a Stark state directed toward the incoming ion. Total capture cross sections from directed Stark states have recently been measured [27], but final-state distributions for these collisions have not yet been observed.

While classical analyses have been used previously to interpret general trends of total capture cross sections, they have seldom been used to predict features of partial cross sections. Experimental measurements of the final-state distribution for capture from ''directed'' states would provide a stringent test (below $\tilde{v}=1$) of the applicability of classical concepts to the interpretation of ion collisions with Rydberg atoms. Furthermore, to our knowledge, the only alternative theory of these collision processes published thus far is the scaled close-coupling calculations of Lundsgaard *et al.* [28–30], and these calculations cannot give meaningful final-state distributions since they are conducted for low-lying principal quantum number states ($n\approx 4$).

Figure 1 displays the classical probability for charge transfer into final states of energy, $E_f=-1/2n_f^2$, resulting from collisions of singly charged ions of impact parameter *b*

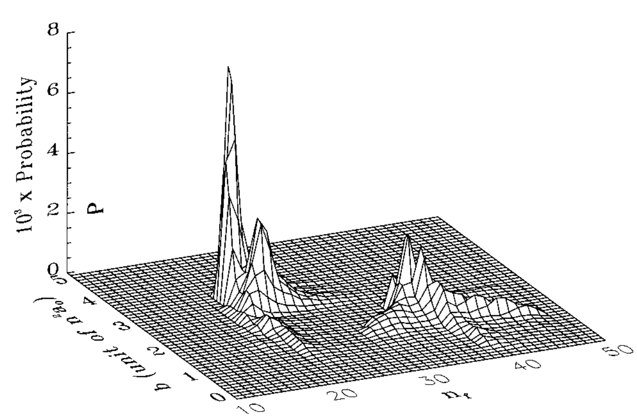

FIG. 1. The charge-exchange probability $(N_{\rm cap}/N_{\rm total})\,b_{\rm max}^2(\pi a_0^2 n^4)$ for an ion-Na(24,23,0,0) collision is plotted versus n_f and initial impact parameter $b(n^2a_0)$.

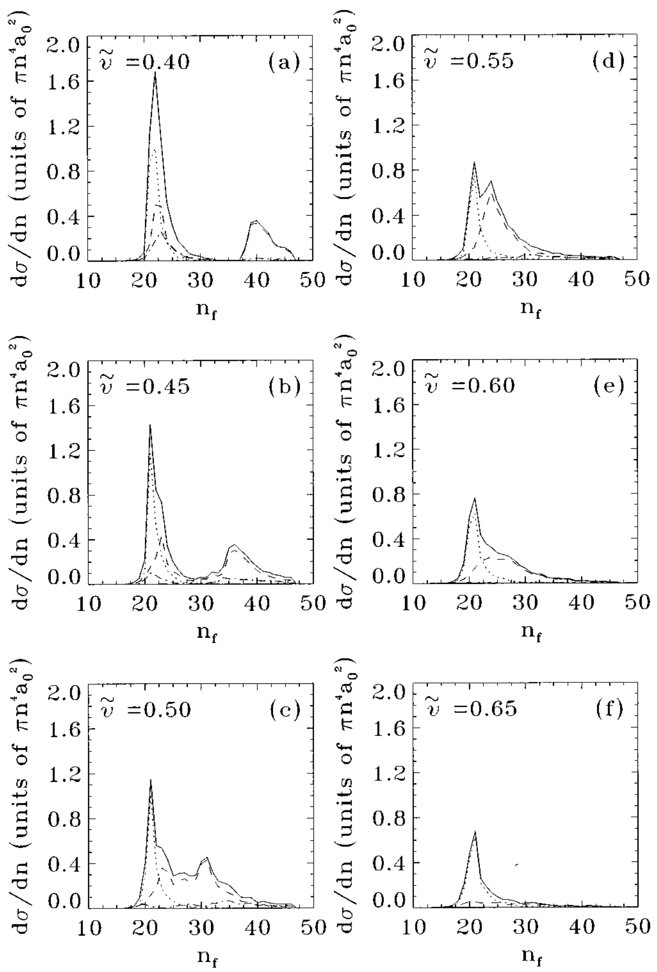

FIG. 2. The charge-exchange probability $(N_{cap}/N_{total})\, b_{max}^2(\pi a_0^2 n^4)$ for an ion-Na(24,23,0,0) collision is plotted versus n_f for $\tilde{v}$ ranging from 0.4 to 0.65. The solid line is the total final n distribution. The short-dashed, long-dashed, and dash-dot lines are the one-swap, three-swap, and five-swap contributions, respectively.

and reduced velocity $\tilde{v}=0.45$, with target atoms consisting of a single electron in an elliptical orbit of energy, $E_i=-1/2n_i^2$, with $n_i=24$ and eccentricity $\epsilon=0.995$. The major axis of the orbit is initially directed toward the incoming ion. This orbit is used to mimic a ''linear'' Stark state with principal quantum number $n_i=24$ and $n_1=23$, $n_2=0, m=0$ [31]. The phase of the electron in its orbit is randomized in accordance with Kepler's equation, as is standard in Monte Carlo simulations [32]. (Details of our numerical methods can be found in earlier works [13–15].)

There are two contributions to capture from upstream-directed Rydberg states apparent in Fig. 1. Large-impact-parameter collisions ($b>2.5n^2a_0$) result in near-resonant electron transfer, with the final binding energy comparable to the target binding energy, $n_f\approx n_i$. However, at smaller impact parameters there is a second contribution to charge transfer, resulting in orbits with $n_f\approx 2n_i$. We will demonstrate below that this second charge-transfer process is three-swap in nature, and that it vanishes when rotational coupling is artificially removed from the classical calculation.

Figure 2 illustrates the dependence of the predicted final-

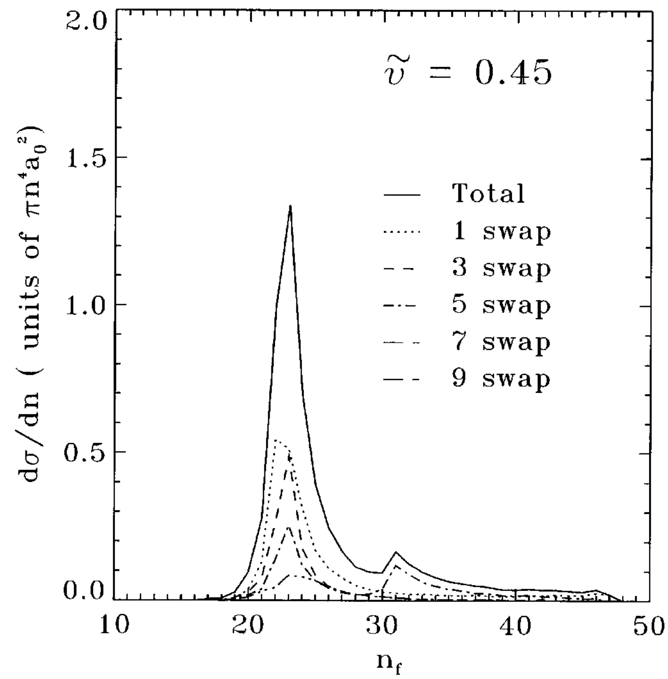

FIG. 3. The charge-exchange probability $(N_{cap}/N_{total})\, b_{max}^2(\pi a_0^2 n^4)$ for an ion-Na(24,23,0,0) collision in the rotational frame with Coriolis terms omitted is plotted versus n_f for $\tilde{v}=0.45$.

state distribution on the collision speed. Note that the two peaks are well separated for the low collision speeds, and that the high-energy peak merges with the resonant peak as the collision speed increases. For reduced velocities, $\tilde{v}<0.5$, the high-energy peak is well resolved, providing a clear signature for future experimental investigations.

The cross sections in Fig. 2 have also been resolved into respective contributions from odd-swap processes. A ''swap'' is counted each time the electron trajectory passes through the midplane between the nuclei. Note that the higher-energy peaks in Fig. 2 are composed almost entirely of three-swap events, while the primary contribution to the resonant peak comes from one-swap trajectories.

The three-swap nature of the relevant trajectories, and the preponderance of these trajectories at small impact parameters, suggest that united-atom rotational coupling plays a large role in the formation of the second capture peak. The rapid rotation of the internuclear line, and accordingly of the midplane between the nuclei, near the closest approach accounts for the additional swaps of the relatively slow moving electron. To test this assertion, we repeated the classical calculations while artificially removing rotational coupling from the calculations. This was accomplished by ignoring Coriolis forces while solving Newton's equations in the rotating internuclear-axis frame. The resulting final-state distribution is shown in Fig. 3, for a reduced ion velocity of $\tilde{v}=0.45$. In the absence of rotational coupling, capture is almost entirely resonant, and there is no indication of a high-n_f peak associated with three-swap capture.

While classical calculations provide order-of-magnitude estimates of total cross sections and indicate general trends of experimental data, there is yet very little evidence that such calculations can be used to predict detailed excitation mechanisms such as that described here. This is particularly true for reduced velocities $\tilde{v}<1$, where classical trajectory Monte Carlo methods have previously been considered as inappropriate [20]. We emphasize that the high-n_f capture peak is a robust feature of the classical calculations and should be seen, at sufficiently small reduced velocities, for any target state directed toward the oncoming ion. Both extremal Stark states and coherent elliptical states of high eccentricity would serve as likely candidates to test this classical prediction.

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