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Electron capture in Ar^+-ion collisions with laser-aligned Rydberg atoms

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We report a study of charge transfer from laser-aligned Rydberg atoms in collisions with Ar^+ ions in the velocity-matching regime. The Rydberg atoms in well-defined magnetic substates were prepared by two-photon laser optical pumping in a weak magnetic field. The results are compared with classical-trajectory Monte Carlo calculations and can also be understood in terms of a simple and "intuitive" picture.

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The electron dynamics in ion-atom collisions is an old and venerable research topic. This, however, does not imply the ready availability of quantitative solutions even for its most fundamental form, the three-body Coulomb problem. In the region of low-lying states requiring a quantum-mechanical treatment, very sophisticated techniques have been developed; in the case of one-electron systems, particle-photon coincidences or (inversely) collisions with laser-prepared partners even allow one to extract quantum-mechanically complete information (for recent overviews, cf., e.g., Refs. [1,2]). The situation becomes more and more complex for higher excited states. Theoretically, particularly in the case of Rydberg atoms, the large level density forbids a quantum-mechanical treatment altogether and one has to resort to classical methods; experimentally, moreover, the data interpretation suffers from the drastically increasing number of open channels that cannot be fully resolved by selective field ionization (SFI), most commonly applied in Rydberg experiments. As a result the quantitative understanding of such processes [3] is still far from the sophistication that can be achieved for low-lying states. This is unfortunate, since collisions involving Rydberg states, being not only of fundamental interest, are also of considerable importance in neighboring fields of physics; for example, surface physics (cf., e.g., Ref. [4]).

In order to obtain more detailed information on such highly excited collision systems, we have recently extended the laser optical pumping technique used in our laboratory [5] to the preparation of Rydberg states. This allows us to control not only the initial n,l quantum numbers, but also (by a suitable choice of the laser polarization) the magnetic substate m. The role of m in Rydberg collisions is a long-standing problem; e.g., for relative collision velocities $V=v/v_e>1$ (v is the collision velocity, v_e the classical electron velocity) Kohring, Wetmore, and Olson [6] suggested that electron capture should be particularly efficient if the Rydberg electron moves parallel to the collision plane. However, no experimental verification of such effects is yet known. In the work to be reported here we present a systematic study of such an initial-state alignment (i.e., magnetic-substate dependence) in ion–Rydberg-atom charge exchange. Experiments as well as classical-trajectory Monte Carlo (CTMC) calculations show that the influence of the initial Rydberg electron alignment on the cross section depends strongly on the collision velocity. In addition, the classical nature of the collision system suggests a simple model that allows a quick assessment of the important physics in an otherwise numerically complex problem.

The process under study is

$$\mathrm{Na}(nlm)+\mathrm{Ar}^+\rightarrow\mathrm{Na}^+ +\mathrm{Ar}^* \ ;$$

the Na target is in a fully determined pure state. The experimental setup will be described here only briefly; a detailed account will be given in a separate paper. A well-collimated sodium beam ($\lesssim 3°$) is intersected at right angles with an Ar^+-ion beam produced in a duoplasmatron source. Photons with wavelengths of around 589 and 410.5 nm, respectively, emitted from two cw argon-ion–dye laser combinations (Coherent) are used to excite the Na atoms via the $3p$ intermediate state to final (nlm) states; the principal quantum numbers lie between $n=20$ and 28. The projectile energy ranges from 0.5 to 5.0 keV. Due to its axial symmetry, the experiment is insensitive to the *sign* of m.

A homogenous magnetic field of 75 G, parallel to the primary ion beam direction, is produced in the collision area by an external pair of Helmholtz coils. At this field strength the Na nuclear spin $I=\frac{3}{2}$ is not fully decoupled from the electronic angular momenta of the $3S$ and $3P$ states (Zeeman regime).

The 589-nm laser frequency is locked to the $3\,^2S_{1/2}\bar{F}=2$, $\bar{M}_F=2\rightarrow 3\,^2P_{3/2}$, $F=3$, $M_F=3$ transition by an external stabilization oven. The blue laser light ($\sim$410.5 nm) transfers some of the $3\,^2P_{3/2}$ population into nd Rydberg states ($n=20$ to 28); in this region of n, the nuclear and electronic spins are fully decoupled from the orbital angular momentum at $B=75$ G (Paschen-Back region). The preparation of pure m_l states is achieved by selecting appropriate laser frequencies and polarizations. Because of the lack of an appropriate feedback signal for stabilization, the laser is continuously scanned around the maximum of the desired Rydberg transition. This procedure gives very reproducible results during an entire experimental run. The modification of the initial Rydberg ensemble by the ambient blackbody radiation (room temperature) has been estimated and found to be negligibly small during the typical experi-

mental time scales ($\sim 1\ \mu s$).

The target Rydberg atoms as well as the charge-exchanged projectiles are detected by selective field ionization. To this end the target atoms are ionized by a pulsed electrical field applied to the collision region; the ions are extracted at an angle of 45° with respect to the magnetic-field axis and detected by a tandem channel plate. Varying the SFI field strength between 0 and 2 kV/cm, the diabatic and the adiabatic parts of the SFI spectra [7] are clearly resolved. They are evaluated taking account of the detector angle with respect to the quantization axis (given by the magnetic field), and allow an independent check of the target n,l,m population. Good agreement is found with the results of the optical excitation calculations; typical Rydberg target densities are 10^3 to 10^4 cm^{-3}. The charge-exchanged highly excited projectiles are ionized within an inhomogenous permanent electrical field encountered roughly 1 μs after the collision region. In this way, the weak charge-exchange signal (appearing at an energy different from the primary ion energy) can be well separated from the strong primary beam by means of a parallel plate energy analyzer; in addition, variation of the electric-field strength allows a rough discrimination of final n states.

The experimental results are displayed in Fig. 1(a). We show the ratios of total cross sections σ_m for capture

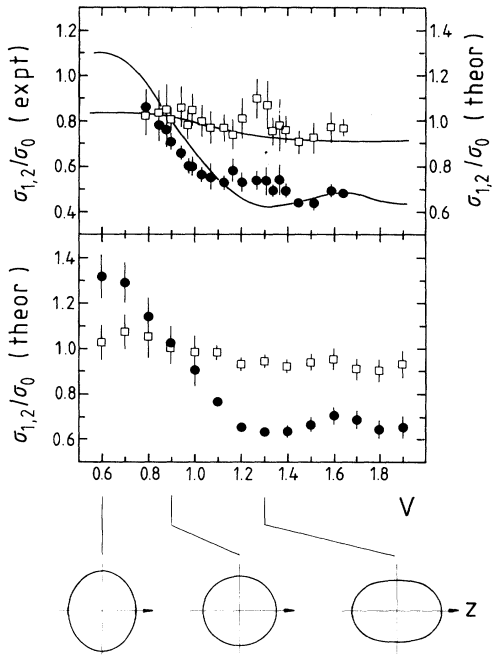

FIG. 1. Charge-transfer cross section ratios σ_1/σ_0 (open symbols) and σ_2/σ_0 (closed symbols) from initial nd Rydberg states with $m=0,1,2$, respectively. (a) Experimental data (left-hand scale); the full line (right-hand scale) is an eye fit to the theoretical data in part (b) of this figure; (b) Results of CTMC calculations. At the bottom of the figure, we show for three selected V values the squares of the $|m|=0,1,2$ wave functions (angular parts), weighted by the respective cross section $\sigma_{0,1,2}$; they may be viewed as initial atomic shapes preferentially involved in the charge-exchange process.

from initial Rydberg states with magnetic quantum number $|m|$ $(=0,1,2)$ as a function of relative collision velocity $V=v/v_e$. The V range is covered by variation of v as well as v_e (i.e., preparing initial states with different n). Apparently, at high collision velocity $V>1$ capture occurs predominantly from low-m states, in qualitative agreement with the expectations of Kohring, Wetmore, and Olson [6], while at low $V<1$ the situation tends to become inverse.

For a quantitative theoretical description of these results, we have performed classical-trajectory Monte Carlo calculations. The code used has originally been developed by Olson [8]. We have implemented the preparation of the initial m states in the following way: Instead of generating randomly oriented Kepler orbits, the classical values of orbital angular momentum and orientation were selected by the condition

$$m \le m_c < m + (l_c - 1) ,$$

where m and l are the correct (quantum-mechanical) magnetic and orbital quantum numbers, respectively; the corresponding classical values chosen in the calculations are labeled by the index c. The calculations were performed for the velocity regime $0.6 \le V \le 2.0$. Each data point represents an average over at least 10^4 trajectories. The resulting cross-section ratios are also shown in Fig. 1.

The agreement between theory and experiment is quite acceptable, although we note an approximately 20% shift of the absolute scales. At present, its origin is unclear. It may point to a deficiency of classical calculations for such fairly low-l states. On the other hand, an experimental problem cannot yet be excluded which is related to the difficult SFI analysis of Rydberg states [3] in the neutralized projectile, particularly if a range of m states is involved. Unfortunately, little is known about this final m distribution; calculations of MacKellar and Becker [9] indicate that it may be rather wide, depending on the initial target state. So far, we did not observe any systematic influence of the initial principal quantum number n on our results; nevertheless, we will extend further experiments to somewhat higher n since the correspondingly higher-n final states are expected to be less sensitive to the details of the SFI detection technique.

Monte Carlo calculations are numerically involved. However, basic principles of charge exchange suggest a simple model that can qualitatively explain the above results, and at the same time allows us to directly visualize the relevant physics; this is aided by the classical situation encountered in Rydberg collisions. The two ingredients are the maximum spatial overlap of the wave functions in the initial target and the final projectile state, i.e., the geometrical cross section, and the additional translational momentum that the electron has to acquire when changing from the stationary target to the moving projectile. The latter (together with the decreasing interaction time) is responsible for the steep charge-exchange cross-section decrease [3] at $V>1$; in a quantum-mechanical formulation, this is usually accounted for by "electron translation factors" (ETF's; for a discussion of various possible forms cf., e.g., Ref. [10]). This

picture immediately suggests that at $V>1$ electron transfer is favored if the initial electron momenta in the target contain components *parallel* to the projectile motion, i.e., particularly in the case of low m states. Indeed, the expectation value $\langle k_z^2 \rangle$ (k_z, the d-Rydberg electron momentum projected onto the primary ion beam direction z, and given in units of $1/n^2$) can easily be calculated to be 0.52, 0.43, and 0.14, respectively, for the $m=0,1,2$ initial states; thus σ_1/σ_0 and σ_2/σ_0 are expected to be slightly less than 1 and clearly greater than 1, respectively, in agreement with Fig. 1. At intermediate velocities $V\approx 1$, the velocity mismatch between the initial Rydberg electron and the moving projectile decreases; correspondingly, ETF's are known to be of decreasing importance, and thus also the role of matching momenta k_z. The geometrical extension of the charge cloud will be therefore of increasing relative influence; higher-m states become favored, extending out to larger distances ρ from the target nucleus perpendicularly to the impact direction. Since for hydrogenic states $\langle \rho^2 \rangle \sim 1-\langle k_z^2 \rangle$, σ_1/σ_0 and σ_2/σ_0 become at $V<1$ slightly and clearly greater than 1, respectively, as also seen in Fig. 1. At very low $V \ll 1$, ETF's and thus the role of matching momenta k_z are insignificant; furthermore, the long duration of the initial part of the collision allows the electron motion to fill the entire accessible phase space before the electron can pass over the Coulomb barrier between target and projectile. This weakens the influence of an initial alignment; as a result, at very low V we would expect the cross-section ratios to approach unity.

In summary, on the basis of a two-photon laser-optical pumping experiment, we have performed a study of alignment-dependent charge exchange involving ion–Rydberg-atom collisions. Although the initial l values are low, classical (CTMC) calculations give a good account of the alignment dependence. In addition, a simple model is able to qualitatively predict the results without the need of elaborate calculations. We want to point out that in case of capture from low-n states around $V\approx 1$ the velocity-matching concept is currently investigated in great detail (cf. [11–13]); qualitative velocity-matching considerations (as also applied above) or, more quantitatively, the collisionally accumulated phases explain the sensitivity of the charge-exchange process to the initial direction of electron motions ("orientation") relative to the projectile trajectory. Similar refinements may also be expected for Rydberg collisions from corresponding more detailed studies. Such work should also shed further light on the still open problem of final states in collisions between atoms and highly charged ions (cf., e.g., Refs. [14,15]) which may in some respects be regarded as an "inverse" of the systems studied here.

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[1] E. E. B. Campbell, H. Schmidt, and I. V. Hertel, Adv. Chem. Phys. **72**, 37 (1988).

[2] R. Hippler and H. O. Lutz, Comments At. Mol. Phys. **28**, 39 (1992).

[3] K. B. MacAdam, L. G. Gray, and R. G. Rolfes, Phys. Rev. A **42**, 5269 (1990), and references therein.

[4] See, e.g., H. J. Andrä *et al.*, in *Physics of Electronic and Atomic Collisions,* edited by W. R. Gillivray, E. McCarthy, and M. C. Standage (Hilger, Bristol, 1992), p. 89.

[5] K. Finck, Y. Wang, Z. Roller-Lutz, and H. O. Lutz, Phys. Rev. A **38**, 6115 (1988); Z. Roller-Lutz, K. Finck, Y. Wang, and H. O. Lutz, Phys. Lett. A **169**, 173 (1992).

[6] G. A. Kohring, A. E. Wetmore, and R. E. Olson, Phys. Rev. A **28**, 2526 (1983).

[7] T. F. Gallagher, L. M. Humphrey, W. E. Cooke, R. M. Hill, and S. A. Edestein, Phys. Rev. A **16**, 1098 (1977).

[8] R. E. Olson, J. Phys. B **13**, 483 (1980); and private communication.

[9] A. D. MacKellar and R. L. Becker, in *Physics of Electronic and Atomic Collisions,* edited by J. Eichler, W. Fritsch, I. V. Hertel, N. Stolterfoht, and U. Wille (Springer, Berlin, 1983), p. 660.

[10] J. S. Briggs, in *Fundamental Processes in Energetic Atomic Collisions,* edited by H. O. Lutz, J. S. Briggs, and H. Kleinpoppen, Vol. 103 of *NATO Advanced Study Institute Series B: Physics* (Plenum, New York, 1983), p. 421.

[11] E. E. B. Campbell, I. V. Hertel, and S. E. Nielsen, J. Phys. B **24**, 3825 (1991).

[12] J. C. Houver, D. Dowek, C. Richter, and N. Andersen, Phys. Rev. Lett. **68**, 162 (1992); and unpublished.

[13] Z. Roller-Lutz, Y. Wang, K. Finck, and H. O. Lutz (unpublished).

[14] See, e.g., H. Cederquist *et al.*, in *Physics of Electronic and Atomic Collisions* (Ref. [4]), p. 391.

[15] J. Pascale, in *Physics of Electronic and Atomic Collisions* (Ref. [4]), p. 401.