n Dependence of *l*-Changing Collisions between He^+ Ions and Na

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l-changing collisions were observed in a crossed He^+-ion/Na–Rydberg-atom beam experiment. Transitions $nd \rightarrow n(l \geq 3)$ induced in Na by ion impact at 450 and 600 eV were studied for $n = 20–34$. Cross sections vary approximately as n^5 and have magnitudes of order 10^8 Å^2, a few hundred times the geometric cross section of the Rydberg atoms.

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The mean radii $\bar{r} \approx n^2 a_0$ of Rydberg atoms, their binding energies $E_n \approx -13.6/n^2$ eV, and their close spacing of energy levels $\Delta E \propto n^{-3}$, as well as other properties,[1] lead to the expectation that collision cross sections for both bound-state and continuum processes may have unparalleled magnitudes at high principal quantum numbers n. The n dependence of Rydberg-state collision cross sections is an important matter for laser modeling, plasma fusion, astrophysical studies, and basic theory. In collisions of charged particles with Rydberg atoms, ionization and charge-transfer cross sections have been found to be of order $n^4 \pi a_0^2$, where a_0 is the Bohr radius, i.e., they approximate the geometrical cross section.[2-4] In collisions of neutral perturbers with Rydberg atoms, however, cross sections for collisions that change the orbital angular momentum l rise as n^4 only to some n_0 that depends on the polarizability of the perturber, and fall for higher n.[5,6]

Percival and Richards[2] predict theoretically that l-changing cross sections in Na(nl)–charged-particle (ion) collisions should be maximum when the projectile velocity v_i and Rydberg-electron orbital velocity v_e are nearly equal. For Na $nd \rightarrow nf$ transitions, although the n dependence is not a simple power law, the cross section is calculated to vary approximately as $n^{4.5}$ for 500-eV He^+ projectiles in the range $n = 20–30$. The maximum cross section, i.e., at optimal v_i, varies as $n^{5.3}$ in this range of n. These faster-than-geometric n dependences are in accord with a simple physical argument given by Percival[2] for $n \rightarrow n+1$ transitions that suggests cross sections of order $n^5 \pi a_0^2$.

In this Letter we report an experiment in which l changes in Na(nd) Rydberg states, induced by He^+ impact at $v_i/v_e = 1.3–2.3$, were found to have cross sections that vary approximately as n^5 and magnitudes on the order of 10^8 Å^2.

Our crossed ion-atom beam apparatus is shown schematically in Fig. 1. Not shown are lasers, a Wien velocity filter, and an electrostatic ion-beam switcher. The ion beam of a few hundred nanoamperes at 450 and 600 eV filled a 6-mm aperture at the entrance to a differentially pumped collision region. A thermal Na atomic beam of diameter 4 mm crossed at the center of this region. Na ground-state densities 2×10^8 cm^{-3} were routinely obtained. A N_2-laser–pumped double-dye-laser system provided nearly simultaneous laser pulses (20 Hz repetition rate) at wavelengths suitable for stepwise excitation of Na atoms[7] at the intersection of ion and atomic beams. About 0.1% of Na atoms in a 10^{-2}-cm^3 volume were excited to selected nd states. The lasers were obliquely polarized and populated all m_j sublevels of the Rydberg states. The time interval during which the Rydberg atoms were a useful target for He^+ impact was limited by their 1-mm/μs thermal velocity across the ion beam.

The number of Rydberg atoms present after laser excitation was determined by field ionization. A positive high-voltage pulse ($t_r = 1$ μs), suitably triggered and delayed, was applied to the upper of two parallel plates surrounding the interaction volume. Na^+ ions produced by field ionization were accelerated downward through a grid in the lower plate into an electron multiplier. The time-resolved field-ionization method was used to discriminate among the populated Na Stark sublevels.[8] We varied laser polarizations, ionizing-pulse magnitudes, etc., and found a pattern of sublevel arrival times broadly consistent with Gallagher *et al.*[8] Diabatic behavior in field ionization of d states reported by Jeys *et al.*[9] was also observed, in which ionization of $|m_l| = 2$ sublevels occurred with threshold fields markedly higher than required for ionization of the $|m_l| = 0, 1$ sublevels that follow adiabatic or partly adiabatic paths to ionizing fields. However, unlike Jeys *et al.* who reported clearly diabatic behavior as low as $n = 30$, we found no completely diabatic behavior for $n < 40$.

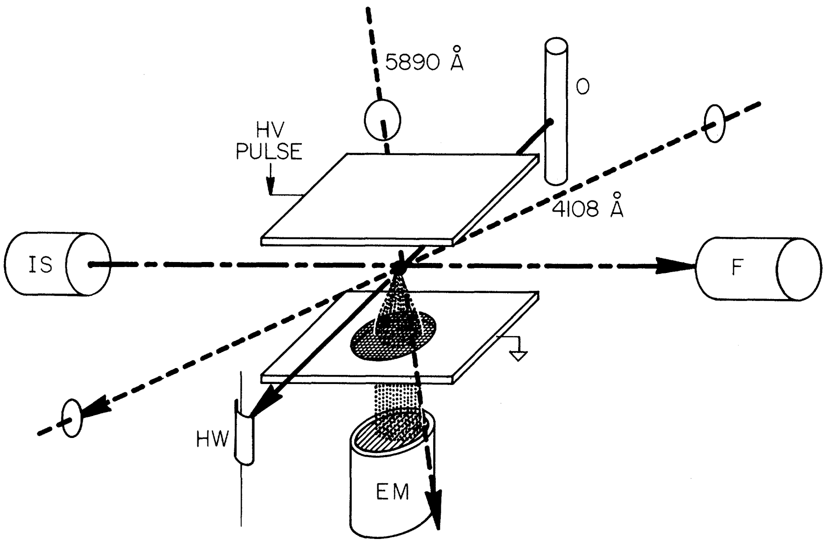

FIG. 1. Schematic diagram of appartus: IS, ion source; O, atomic beam oven; F, Faraday cup; EM, electron multiplier; HW, hot-wire detector. Long and short dashed line, ion beam; solid line, Na beam; dashed lines, laser beams.

To observe ion-Rydberg collisions, we alternately switched the ion beam *on* for 1.5 μs following a number of laser flashes, then *off* during an equal number. When the lasers were tuned to a *d* state, e.g., 28*d*, the resulting field-ionization signals differed markedly in beam *on* and *off* modes, as shown in Fig. 2. In particular, when the ions were gated *on*, the major peak representing all components of the parent *nd* state was diminished as much as 30% in area, and a new peak, corresponding to threshold fields some 80% greater, appeared with area equal to that lost from the main peak. We separately identified the position of this new peak as that of field ionization through diabatic passage to high fields[9] of $|m_l| \geq 3$ components of $f, g, h, \ldots$ states have the same n. (Laser excitation in the presence of a small dc electric field allowed excitation of $l \geq 3$ states. With dc and pulsed fields *of opposite polarity,* production of $|m_l| = 3$ sublevels was enhanced through nonadiabatic rapid passage occurring during the reversal of the net-field direction.) The fractional reduction R in the area of the parent peak represents the probability that a single Rydberg atom undergoes a collisionally induced transition to a state that field ionizes with significantly different threshold. Although this measure includes collisional ionization, charge transfer, and n-changing collisions, the first two have cross sections expected to be roughly $n^4 \pi a_0^2$, whereas the magnitude of the parent peak reduction corresponds to a cross section two orders of magnitude larger. No other collisionally produced n's down to at least $n = 14$ were found. R will not measure, however, transitions to Stark sublevels that field-ionize adiabatically or partly adiabatically, for these will

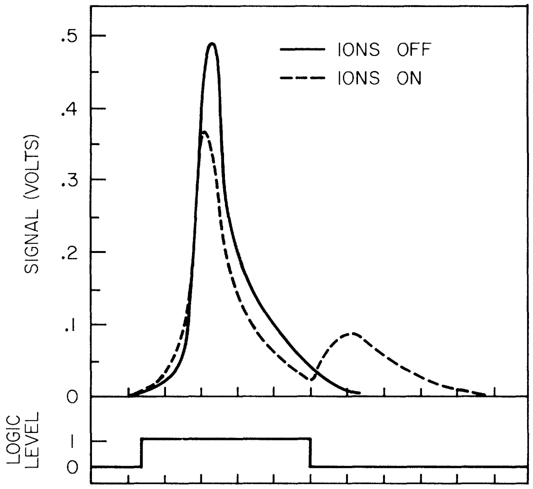

FIG. 2. Time-resolved field-ionization signals, with and without He$^+$-ion beam. The parent Na(28*d*) state is seen at 7.65-μs delay. All $|m_l|$ components fall within the integration window shown. He$^+$ impact at 450 eV causes a depletion of the parent state and the appearance of a new signal at 8.0 μs identified as $|m_l| \geq 3$ components of Na(nl) for $l \geq 3$.

contribute signals within the parent peak. That is, only $\frac{2}{7}$ of the $nd \rightarrow nf$ collisions will be counted, crudely based on statistical weights.

From the definition of a collision cross section we may take

$$\sigma = R(J\Delta t/e)^{-1} \tag{1}$$

for the observable part of the $nd \rightarrow n(l \geq 3)$ cross section induced by ion impact. J is the ion beam current density, Δt is the time of exposure of Rydberg atoms to ion impact before field ionization, and e is the electronic charge. The linear relationship between R and J or Δt in (1) was verified experimentally within the limits of reproducibility of the data.

For the $28d \rightarrow 28(l \geq 3)$ process induced by 450-eV He^+ projectiles, we find $R = 0.210 \pm 0.015$ with $J = 8.5 \times 10^{-7}$ A/cm^2 and $\Delta t = 1.5$ μs. Thus $\sigma \approx 2.6 \times 10^{-8}$ cm^2 = 260 000 000 Å^2. Percival and Richards[2] predict $\sigma = 7 \times 10^{-8}$ cm^2 for Na $nd \rightarrow nf$ in which, of course, all $|m_l|$ components are included. Classical-trajectory Monte Carlo calculations recently performed by MacKellar and Becker[10] give $\sigma = 1 \times 10^{-8}$ cm^2 for the same process and energy. For comparison, $n^4 \pi a_0^2 = 5.4 \times 10^{-11}$ cm^2 at $n = 28$. The reduced velocity $v_i/v_e = 1.9$ for He^+ at 450 eV. Flannery and McCann[11] find that in charged-particle–Rydberg-atom collisions, *non*dipole processes (e.g., $nd \rightarrow nl$, $l > 3$) may be dominant. At this time we cannot distinguish nf states from higher l or rule out the possibility of a chain of successive transitions $nd \rightarrow nf \rightarrow ng\ldots$ induced by several ion impacts, in which the first step serves as a bottleneck. Nonetheless, we feel our result is an order-of-magnitude verification of current theories.

Figure 3 shows R for $n = 21$–34 at 450 and 600 eV. For $n \gtrsim 28$ we could no longer completely time resolve the collisional signal from the $|m_l| = 2$ components of the parent signal, because of the increasingly nonadiabatic behavior of $|m_l| = 2$ sublevels. The leveling off in Fig. 3 suggests that transitions to $|m_l| \geq 3$ are relatively less likely for the d-state sublevels that field ionize along adiabatic paths ($|m_l| = 0, 1$) than for those that exhibit nonadiabatic behavior ($|m_l| = 2$). Over the range $n = 21$–28 at 450 eV the data yield a scaling exponent ($\sigma \propto n^\beta$) $\beta = 5.17 \pm 0.35$. The $n = 20$–27 data at 600 eV yield $\beta = 5.16 \pm 0.15$.

A number of effects were considered that might influence the interpretation of our results. Photons and neutral atoms from the ion source caused no observable effect on the Rydberg atoms. Metastable He^+ would be quenched in the ion optics. He^+ that left the ion source in Rydberg states would radiatively decay or be collisionally quenched or ionized during the 100-cm flight to the collision region. Furthermore, He^+ Rydberg ions are unlikely to induce l changes much differently from $\mathrm{He}^+(1s)$ at the impact paremeters $b \approx 10 n^2 a_0$ effective in the observed process. The space-charge fields in the ion beam were much less than 0.1 V/cm. Application of external fields even two orders of magnitude greater with ions *off* did not produce the observed effects. The experiments were repeated with the ion beam *on* during the flash of the lasers to see whether the presence of ions allowed other states to be laser excited. However, the Rydberg-state excitation spectrum remained unchanged as did the cross-section measurements, indicating that the high-l states were produced by collisions from the directly excited nd states.

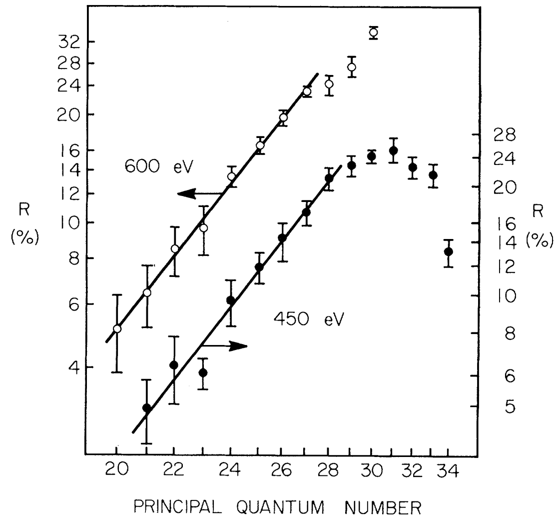

FIG. 3. R, proportional to Na(nd) l-changing cross section, vs n for 450- and 600-eV He^+ impact. Note the vertical offset of left and right logarithmic scales, introduced for clarity. Different ion-beam currents were used at the two energies. Least-squares fits are shown with slopes (exponents) give in the text.

This is believed to be the first experimental observation of a faster-than-geometric n dependence for collisions of Rydberg atoms with charged particles. The enormous excess over geometrical cross sections, indicating effective impact parameters as large as the wavelength of visible light, makes the l-changing process one that could well affect the behavior of many laboratory and astrophysical plasmas. Detailed further studies will be reported in later publications.

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Experimental Confirmation of a Scaling Law for the 1*sσ* Excitation Probability for $Z_1 + Z_2 > 120$, and its Breakdown in Pb + Cm Collisions at Very Small Internuclear Distances

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The excitation probability of the 1$s\sigma$-molecular orbital has been measured as a function of the impact parameter b for several collision systems in the region $129 \leq Z_1 + Z_2 \leq 178$ at the UNILAC. All published impact-parameter data agree with a simple scaling law except those for impact parameters $b \leq 40$ fm in the Pb + Cm system ($Z_1 + Z_2 = 178$).

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In adiabatic collisions between heavy ions Z_1 and atoms Z_2 with $\alpha(Z_1 + Z_2) = \alpha Z \gtrsim 1$, superheavy quasimolecules are formed transiently. Because of the strong Coulomb field of both nuclei the binding energy of the innermost electrons is of the order of the electron rest mass. Such highly relativistic systems exhibit a characteristic and most exciting feature, namely, a tremendous shrinking of the spatial wave functions of the electrons and a corresponding increase of their high-momentum components.

Collision systems with $\alpha Z \gtrsim 1$ have been studied experimentally via K-x-ray emission within the last few years very extensively by measurements of the excitation probability $P_{1s\sigma}(b)$ of the molecular 1$s\sigma$ orbital in encounters with impact parameters b.[1] These experiments confirm indeed that just the relativistic effects lead to high values of