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Enhancement of charge capture from a laser-excited target by highly charged ions

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A charge-transfer experiment has been done in which 63-keV Ar^{7+} was incident on sodium in the ground and first excited states. The electron-capture ratio $\sigma(\mathrm{Na}(3p))/\sigma(\mathrm{Na}(3s))$ was experimentally determined for this system. The charge-capture cross section was observed to increase by a factor of 1.5 for the excited target.

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An area of atomic physics that has received much attention in recent years concerns collisions between ions and laser-excited targets [1–8]. A laser-excited target can be of interest for a number of reasons, including, for example, studies of alignment and orientation [1–8] effects on charge capture. In this work the primary aim is to modify the target binding energy and observe the effect on charge capture by highly charged ions at low collision velocities. Ar^{7+} was chosen as the projectile because this collision system is at the current limit [9,10] of applicability of quantum-mechanical (e.g., coupled-channel) models. Furthermore, Ar^{7+} has no long-lived metastable states that might make capture measurements ambiguous.

In general, classical over-the-barrier models (OBM's) predict capture cross sections to vary inversely with the square of the target binding energy in the velocity regime investigated here (0.25 a.u.). Classical-trajectory Monte Carlo (CTMC) calculations have been made for this system [11], but may be suspect in this velocity regime [12]. Coupled-channel calculations have not been published for this system to our knowledge. The major difficulty with quantum-mechanical calculations for this sort of collision system is that the electron is captured to predominantly high-n states ($n \sim 10$ for this system, according to CTMC calculations). Thus a very large number of states must be included in the basis set, leading to calculation difficulties. The experiment reported here provides a check on current classical models, as well as on future quantum-mechanical calculations.

The details of the experiment are described elsewhere [13]. Briefly, a collimated beam of Ar^{7+}, produced in the Kansas State University electron-beam ion source (EBIS) [14], is crossed at 90° with an effective jet of sodium vapor, produced in a stainless-steel oven heated to approximately 350 °C. The ion beam is then charge-state analyzed with an electrostatic deflector and a two-dimensional (wedge and strip) position-sensitive detector (PSD). The sodium from the jet is collected in a water-cooled trap. A laser, tuned to a $3s \rightarrow 3p$ absorption line in sodium, crosses both the ion beam and sodium jet at 90° in the interaction region. Care was taken to insure that all of the sodium target crossed by the ion beam was exposed to saturating intensities of laser radiation. The laser is linearly polarized parallel to the ion-beam axis.

The excitation system consists of a grazing-incidence dye laser pumped by a copper-vapor laser (CVL). The CVL is a pulsed laser having a nominal pulse length of 25 ns and a repetition rate of 6 kHz. The CVL lases at 511 and 578 nm; the 511-nm line was used to pump the dye laser. The dye-laser linewidth $\Delta\nu_L$ is estimated to be ≤ 1.5 GHz, and the peak intensity is about 0.7 kW/cm^2.

The relevant energy-level diagram for sodium is shown (not to scale) in Fig. 1. Because $\Delta\nu_L$ is smaller than the hyperfine splitting of the ground state, but large compared with the hyperfine splitting of the excited states, four transitions are available for laser excitation:

$$3s_{1/2}, F=1 \rightarrow 3p_{3/2}, F=0,1,2\ , \tag{1a}$$

$$3s_{1/2}, F=2 \rightarrow 3p_{3/2}, F=1,2,3\ , \tag{1b}$$

$$3s_{1/2}, F=1 \rightarrow 3p_{1/2}, F=1,2\ , \tag{1c}$$

$$3s_{1/2}, F=2 \rightarrow 3p_{1/2}, F=1,2,\ . \tag{1d}$$

All four transitions were investigated with similar results, but the transition shown in (1b) is the focus of this work.

A fraction of the dye-laser output was made to be in-

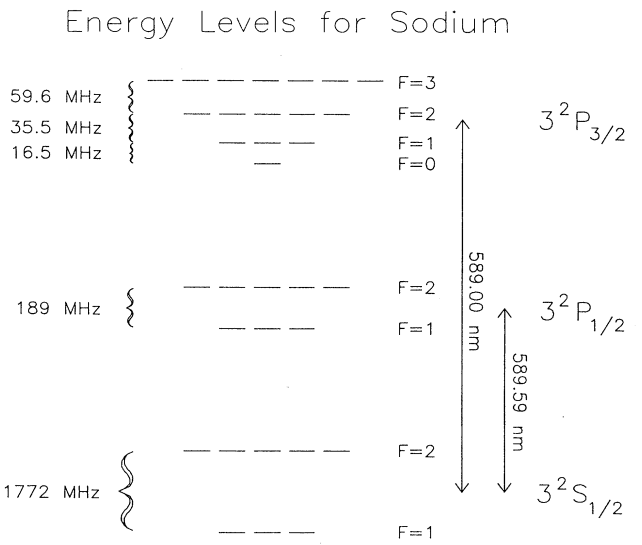

FIG. 1. Energy-level diagram for sodium showing fine and hyperfine splitting (not to scale).

cident on a fast *p-i-n* photodiode. A delayed signal derived from the *p-i-n* diode output was used to start a time-to-amplitude converter (TAC). A fast timing signal from the PSD was used to stop the TAC. The output of the TAC was digitized and sent along with the digitized output of the PSD to a μVAX workstation. The data were taken in list mode and sorted such that only TAC signals corresponding to Ar^{6+}, i.e., the capture channel, were considered. The sorted TAC spectra show the number of charge-capture events in the time intervals before, during, and after the target is excited by the laser pulse.

In order to insure single-collision conditions, the percent capture from ground-state sodium was measured as a function of sodium pressure in the oven. As long as the conditions for effusive flow are maintained, the target density in the interaction region is related to the oven pressure by a constant. The oven pressure was estimated by measuring the oven temperature and using published vapor pressure curves [15]. By extrapolating the pressure-dependence curve to zero pressure, one also obtains an estimate of capture from slits and background gas. This was found to be 1.3% for the series of runs whose results are presented here. For the cross-section ratios reported here, the oven pressure was maintained at 75 mTorr, leading to an estimated target density at 10^{12} atoms/cm^3 in the interaction region.

A TAC spectrum for transition (1b) is shown in Fig. 2. This is a plot of the number of charge-transfer events as a function of time. One can clearly see the rise in charge-transfer events when the laser turns on. The capture rate reaches a roughly constant plateau while the laser is on, and decays exponentially in time when the laser turns off. This is consistent with the expected temporal dependence of the excited-state population. The ratio of cross sections was determined using the following relation:

$$\frac{\sigma_{3p}}{\sigma_{3s}} = \frac{R-1}{f_{\rm ex}} + 1 \ , \tag{2}$$

where σ_{3l} is the capture cross section from Na($3l$), $f_{\rm ex}$ is the fraction of sodium atoms in the excited state, and R is the ratio of charge-capture events with laser on to laser off. The ratio R is first corrected for capture from background gas (as estimated from the above-mentioned pressure-dependence measurements) and then for capture from sodium dimers (at this temperature estimated [15] to be about 1.7%).

In order to use Eq. (2) to evaluate the cross-section ratio, the excited-state fraction must be known. While methods have been developed [16] to directly measure the excited-state fraction in sodium after laser pumping, a different approach was used here. Under saturation conditions, and with a known polarization of the exciting radiation, the calculation of the excited-state fraction is straightforward. As mentioned above, the entire target area "seen" by the projectile beam was exposed to saturating intensities of laser radiation. The question of polarization was a little more difficult to resolve. The target was subjected to laser radiation that was strongly linearly polarized along the projectile beam axis. However, as can be seen in Fig. 2, the artificially long radiative lifetime of the sodium (about 90 ns) shows [17] that radiation trapping is occurring. Under these conditions, there is no guarantee that the target is exposed solely to linearly polarized radiation [18]. Thus part of the uncertainty in the determination of the ratio of cross sections is due to the uncertainty in the excited-state fraction, which in turn is due to the uncertainty in the effective polarization of the exciting radiation field. Excited-state fractions were calculated by solving sets of 36 coupled rate equations for a variety of polarizations. The excited-state fraction varies with time during the 20 ns of laser pumping, with an initial rapid rise to a maximum, followed by a slow decay. The calculated $f_{\rm ex}$, after 10 ns of laser pumping was used with Eq. (2) to determine the ratio of capture cross sections.

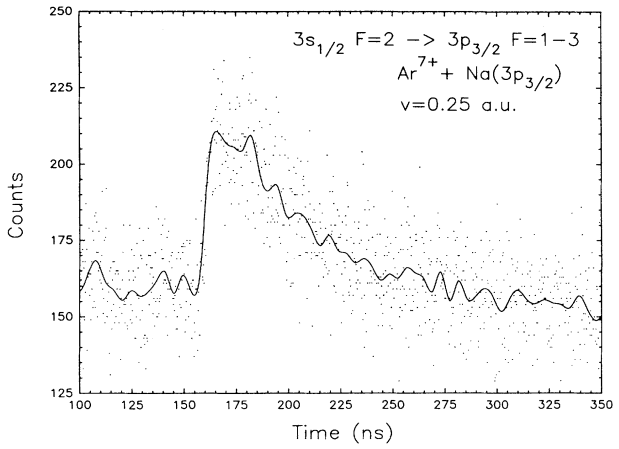

FIG. 2. Number of capture events as a function of time for the $3s_{1/2}, F=2 \rightarrow 3p_{3/2}, F=1,2,3$ transition. The points are the raw data, while the solid line represents an average over 16 adjacent points. In this figure the laser turned on and off at approximately 155 and 180 ns, respectively.

The uncertainty in the excited-state fraction estimated from these calculations was used in the determination of overall experimental error. For transition (1b) the calculated excited-state fraction was 0.40 for linearly polarized light, 0.39 for circularly polarized light, and 0.43 for unpolarized light. Thus the uncertainty in $f_{\rm ex}$ was about 7.5%. As an aside, it should be mentioned that the target lifetime enhancement due to radiation trapping allowed a second independent estimate of target density [17], which was consistent with the 10^{12} atoms/cm^3 estimated from vapor pressure curves.

Using Eq. (2), the TAC spectrum of Fig. 2 yields a cross-section ratio of 1.53 ± 0.28 for the transition shown in Eq. (1b). The error estimate includes statistical error as well as the aforementioned uncertainty in excited-state fraction. CTMC and OBM calculations both give cross-section ratios of 2.1 for this system. The disagreement between experiment and these models is greater than the experimental error bars, but may be due to the inadequacy of the hydrogenic potentials used in the calculations.

In summary, an enhancement to the capture cross section was observed for slow, highly charged ions on a sodi-

um target when the target electron's binding energy was reduced. This is in agreement with classical models. However, on an absolute scale there is some discrepancy between theory and experiment. Experiments are being due to determine absolute cross sections for capture from both ground and excited states of sodium by Ar^{7+} over a range of collision velocities. In addition, experiments will be extended to higher-charge-state projectiles in order to examine the capture cross-section dependence on projectile charge.

All of the CTMC calculations were done using a computer code written by Dr. N. Toshima, which was made available to us by him and through the efforts of Professor R. Shingal and Professor C. D. Lin. We would like to thank these colleagues for their help. We would also like to acknowledge many helpful discussions with Professor R. Singal and Professor L. Weaver. This work was supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Energy Research, U.S. Department of Energy.

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