Lyman-α emission in collisions of H^+ ions with Na(3s) and laser-excited Na(3p) atoms

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We report on an experimental study of Lyman-α emission from H(2p) capture processes in collisions between H^+ ions and Na(3s) as well as laser-excited aligned Na(3p) atoms. The impact energy lies between 0.5 and 10 keV. We observe a strong enhancement of capture from Na(3p) over capture from Na(3s), in agreement with recent calculations by R. Allan *et al.* and W. Fritsch, but at variance with earlier experimental and theoretical results (V. S. Kushawaha *et al.* and M. Kimura *et al.*).

I. INTRODUCTION

In the course of the development of modern atomic physics, the field of atomic collisions has always been an important research subject. Not surprisingly, from this long history a high level of understanding has evolved; in certain selected cases involving simply structured collision systems the description can even be quantum-mechanically complete. It is interesting to note, however, that in the great majority of cases such studies involved collisions between projectiles and atoms in isotropic ground-state configurations. Only within approximately one decade, has laser optical pumping provided an efficient experimental technique to break this special symmetry, thereby triggering a rapidly increasing activity in this field. Orbital angular momentum preparation illuminates the role of "shape" and "rotation" of the initial atomic state, and may also provide data relevant to more applied fields in which excited-state atom collisions play a significant role (e.g., fusion technology, or laser development). Most of this work has concentrated on low-energy interactions of photons, electrons, atoms, and molecules with state-prepared atoms (for overviews cf., e.g., Ref. 1–3). Rather little is known, however, about higher interaction energies. Recently, Bizeau *et al.* (cf. Ref. 4) observed photoionization of laser-excited Na and Ba with synchrotron light ($h\nu$=20–150 eV); Schneider *et al.*[5] reported autoionization spectra following interaction of 1-MeV He^+ ions with Na(3p) $^2P_{3/2}$ atoms; Kushawaha *et al.*[6] have studied Lyman-α radiation emitted from H(2p) in collisions between H^+ ions and laser-excited Na atoms in the energy range 1–600 eV. In the latter work the reaction

$$H^+ + Na(3s) \rightarrow H(2p) + Na^+ \quad (1)$$

is endothermic by 1.74 eV, while

$$H^+ + Na(3p) \rightarrow H(2p) + Na^+ \quad (2)$$

is slightly exothermic by 0.37 eV. Due to this near-resonant-level configuration in the second reaction, a pronounced cross-section increase should be expected for capture from Na(3p) compared to capture from Na(3s). It has been speculated that this high-cross-section ratio $\sigma(3p)/\sigma(3s)$ near 1-keV impact energy [the cross sections referring to H(2p) capture from Na(3p) and Na(3s), respectively] may be used to produce inversion in a Lyman-α laser.[7] In these early experiments, an enhanced Lyman-α production up to 30-eV collision energy was observed when the laser line was tuned to the Na D transition; on the other hand, contradictory evidence exists from theory: molecular orbital (MO) calculations by Kimura *et al.*[8] are in agreement with these experiments, while later calculations by Allan *et al.*,[7] based on atomic and molecular orbital (AO and MO) expansions, predict a continuing and strongly enhanced $\sigma(3p)$ cross section over $\sigma(3s)$ up to impact energies of a few keV.

As an extension our "complete experiments" on ground-state ion-atom collisions[9] to collisions involving state-prepared partners, we recently began a more systematic study of the H^+-Na system in the keV energy range. In this paper we report on the cross-section measurements for Lyman-α photon emission in H^+ collisions (energy range 0.5–10 keV) with Na(3s) and Na(3p) atoms.

II. EXPERIMENT

The experiment is shown schematically in Fig. 1. A proton beam from a Colutron ion source is accelerated to an energy in the range 0.5–10 keV. After mass selection in a Wien filter, and collimation in a differentially pumped drift tube, the ion beam is injected into the target chamber. The target chamber is free of electric fields to avoid quenching of H(2s); the earth's magnetic field is compensated with Helmholtz coils to values smaller than 50 mG. In the interaction region, the ion beam of 3 mm diameter is crossed at right angles by a Na-atom beam and a laser beam. The Na beam is produced in a two-stage oven to ensure a small dimer content. After skimming, it is collimated to a divergence below 0.2°; in the target zone, its extension along the laser beam direction is 0.5 mm only (in a few experiments it was 0.2 mm; no difference was noted), the atom density being about 10^9–10^{10} cm^{-3}. The laser beam is emitted from a single-mode cw linear dye laser (Coherent, Inc. CR 599) pumped by a (Coherent, Inc.) argon-ion laser. Its frequency is tuned to the Na(3s) $^2S_{1/2}(\bar{F}=2) \rightarrow$

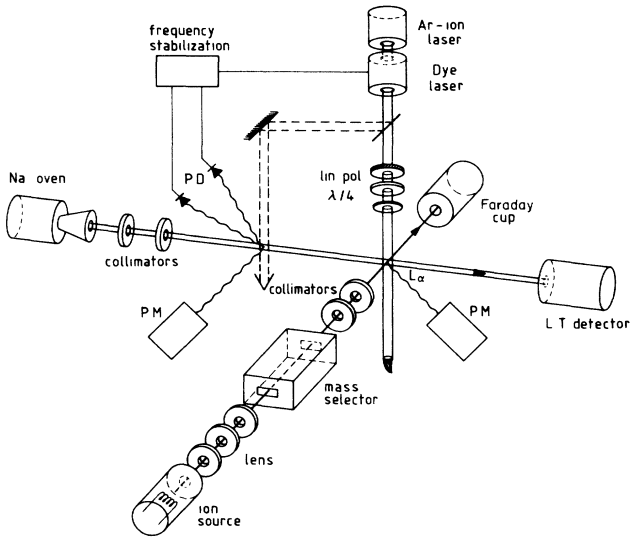

FIG. 1. Experimental setup (schematic).

$Na(3p)\,^2P_{3/2}(F=3)$ transition; the linewidth is about 1 MHz; the laser power in the target region can be varied up to 30 mW/cm^2. A small fraction of the laser beam is used for frequency stabilization, as described in an earlier paper.[10] This feature as well as the good collimation and spatial definition of the Na beam are important to ensure controlled and constant overlap between the pumped Na-beam zone and the ion beam in the interaction region. Lyman-α radiation (121.6 nm) emitted from capture-produced $H(2p)$ is detected in a solar-blind multiplier at approximately the "magic angle" (54.7°) relative to the ion-beam axis to avoid the influence of a possible anisotropic emission on the cross-section measurement.[11] The photomultiplier (MgF_2 window) has a sensitivity window between approximately 115 and 200 nm; it views a length of ion beam around the interaction region of about 3 mm.

III. RESULTS AND DISCUSSION

In order to measure cross sections for capture from $Na(3s)$ and $Na(3p)$, the laser beam and the Na beam were mechanically chopped, and the photon signal detected in synchronization with the choppers. Data were taken by repeatedly alternating laser-on and laser-off as well as Na beam-on and beam-off conditions. In the case of "laser on," both species, sodium in the $3S_{1/2}$ and the $3P_{3/2}$ state, are present. The "effective" total cross section is then given by $f\sigma(3p)+(1-f)\sigma(3s)$ where $\sigma(3s)$ and $\sigma(3p)$ are the cross sections for reactions (1) and (2), and f is the fraction of Na atoms in the $3P_{3/2}$ state. We are assuming here that the excitation mechanism prepares a statistical mixture, rather than there being a coherent superposition of atoms in the ground and excited states. Furthermore, since the $P_{3/2}$-$P_{1/2}$ energy difference is only approximately 2 meV, cross sections at our impact energies are generally assumed to be independent of j (cf. also Ref. 7); hence it is justified to use the term $Na(3p)$ for the initial state. In order to determine the total cross section for $H(2p)$ capture from $Na(3p)$, one must know f.

For an "ideal" optical pumping situation, this quantity is given by the "saturation curve" ("ideal" in the sense that all atoms with $\bar{F}=2$ in the interaction zone participate equally in the pumping process, i.e., $\frac{5}{8}$ of all Na atoms). The saturation curve represents the fluorescence intensity as a function of laser beam intensity. At low laser power, it shows a linear behavior and levels off at high laser power, when the population is shared equally between the $\bar{F}=2$ ground state and the $F=3$ excited state (roughly beyond 30 mW/cm^2). Unfortunately, the situation is complicated by various effects: at very low laser intensities, stationary pumping conditions are not reached in the finite interaction zone; at high intensity, effects of power broadening and trapping cause deviations from the calculated curve; in addition, only a fraction f_p of the $\bar{F}=2$ ground-state atoms will participate in the pumping process, mainly because for a portion of the Na beam the absorption profile will be Doppler-shifted out of resonance due to the finite collimation. This fraction f_p can be determined through the momentum transfer per absorbed photon h/λ which results in a recoil angle $\gamma_0=h/Mv\lambda$ (λ is the photon wavelength; M and v the mass and velocity of the sodium atoms). This technique is commonly used in excited-atom collision studies (cf., e.g., Refs. 2 and 12, and references therein). An atom will undergo an average number N of absorption and spontaneous decay processes in the laser beam. It will therefore suffer a net momentum transfer Nh/λ from the absorbed photons, and the net recoil angle will be $\gamma=N\gamma_0$. Thus the atom beam subtending an angle γ with the initial direction has participated in the optical pumping process. In our experiment, the Na atoms are detected in a Langmuir-Taylor detector at a distance of about 125 cm from the interaction region. In Fig. 2 laser-off and laser-on Na-beam profiles are shown. The portion f_p of atoms which participate in the optical pumping process is given by the ratio of deflected and undeflected beam; their excited-state fraction, in turn, is then obtained for each individual experimental condition from the value of the saturation curve at the appropriate laser power. The pump rate f is then the product of both quantities; typical values lie between 13% and 18%. Note that the Na-beam collimation is always very narrow (0.5 mm or below, parallel to the laser-beam direction). Therefore, the recoil deflection measurement has been performed simultaneously with each cross section measurement; in addition, the portion f_p of atoms participating in the pumping process is high.

Although the laser light is fully polarized, fine and hyperfine interaction causes the excited electron cloud to precess; as a result, $3p_x$, $3p_y$, and $3p_z$ states are populated (z being the axis along the laser polarization direction). In accordance with Fischer and Hertel,[13] we assumed the stationary-state populations to be given by

$$[Na(3p_x)]:[Na(3p_y)]:[Na(3p_z)]=0.22:0.22:0.56\ . \qquad (3)$$

As has been mentioned above, an actual experiment is complicated by several effects which may also cause devi-

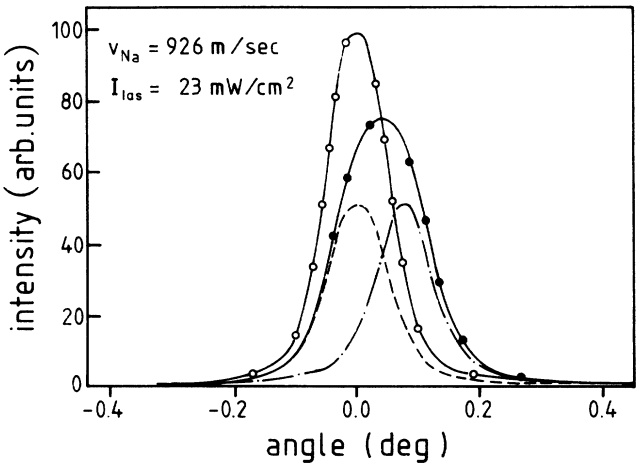

FIG. 2. Beam profiles: –○–, without laser; –●–, with laser; — — —, ground-state beam; —·—·—·, excited-state beam.

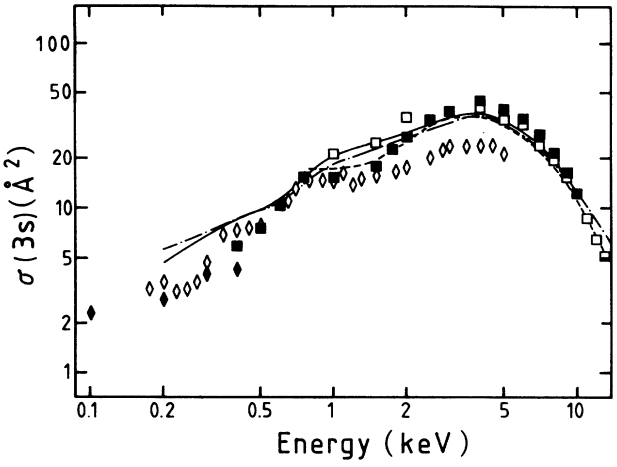

FIG. 3. Experimental Lyman-α emission cross sections and theoretical cross sections for single-electron capture into H(2p) for impact of H^+ on Na(3s). Experimental data: ■, this work; □, Aumayr and Winter (Ref. 16); ◇, Nagata and Kuribara (Ref. 17); ♦, Kushawaha *et al.* (Ref. 6). Theoretical data: ——, Fritsch (Ref. 15); —·—·—·, Allan *et al.* (Ref. 7); — — —, Shingal *et al.* (Ref. 18).

ations from Eq. (3). As will be shown below, the corresponding influence on the experimental cross sections will be small. In any case, since in our cross section measurements the laser power did not exceed 25 mW/cm², and the Na atom density remained below 10^{10} cm^{-3}, we believe we have avoided such complications (cf. also Ref. 14).

The σ(3s) cross section. To put the measured σ(3p) cross-section data on an absolute scale, we first obtained the Lyman-α emission cross section σ(3s) for reaction, Eq. (1). We keep the Na target density constant and determine relative cross sections for impact energies between 0.5 and 10 keV. The data points at and above 1 keV were obtained with H^+ projectiles; at lower energies D^+ projectiles were used for reasons of beam intensity. Since the scattering angles for the charge exchange process considered are quite small (typically below 1°), trajectory effects are negligible, the important quantity being the impact energy per mass unit. All data were taken under single-collision conditions with proper correction for background contributions obtained from Na beam-on and beam-off measurements. The cascade contributions were estimated from the theoretical work of Fritsch[15] for capture into H(3s) and H(3d) states, by taking into account their lifetimes and the geometry of our experiment (in particular, the length of interaction region viewed by the Lyman-α photomultiplier). From these considerations it turned out that in the observed impact energy range cascade contributions to Lyman-α emission for capture from Na(3s) never exceed 2%. The Lyman-α emission cross sections from these measurements were made absolute by normalization to the theoretical results of Fritsch[15] at 10 keV impact energy. In Fig. 3 the resulting cross sections are displayed and compared with available experimental and theoretical data. We find reasonable agreement between our experimental results and those of Aumayr and Winter;[16] those from Nagata and Kuribara[17] show a similar energy dependence, while the absolute values are generally lower. The agreement with calculations of Fritsch,[15] Allan *et al.*,[7] and Shingal *et al.*[18] is good.

The σ(3p) cross section. In order to measure the cross section for capture from Na(3p), we injected the laser beam into the H^+-Na interaction region as described above. The linear polarization direction was chosen perpendicular to the H^+-beam direction. The experimental Lyman-α rate was corrected for cascade contributions by using theoretical results of Fritsch[15] for capture from the Na(3p) into the H(n=3) state at an energy of 1 keV; the cascade correction amounted to about 5%. The same value was applied at all impact energies; at smaller energies this probably overestimates the cascades; at higher energies they may be somewhat more important, but they still remain a small correction to the final result. It should be emphasized that the experimental σ(3p) cross sections are weighted averages for capture from the initial p_x, p_y, p_z states of Na(3p) which are unequally populated ("aligned"), cf. Eq. (3). To compare with theory, we have calculated these weighted cross sections from the data given by Allan *et al.*[7] and Fritsch.[15] As can be seen in Fig. 4(a), the measured cross-section enhancement shows good agreement with both calculations; it is, however, at variance with the earlier work of Kushawaha *et al.*[6] and Kimura *et al.*[8] The σ(3p) cross-section behavior, as obtained by applying this enhancement factor to the σ(3s) cross section, is shown in Fig. 4(b). As expected for a near-resonant charge-exchange process, it has a rather flat energy dependence at low impact energies, before it falls off at higher energies.

Finally, it may be interesting to note that, for pumping with linearly polarized light, the cross section σ(3p) as it is determined in an experiment is rather insensitive to the

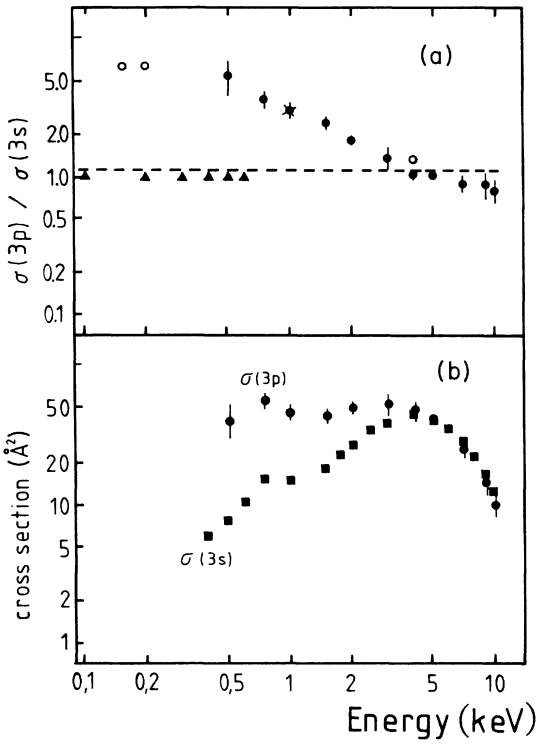

FIG. 4. (a) Enhancement of Lyman-α production for H($2p$) capture from Na($3p$) over Na($3s$); laser light polarization perpendicular to the projectile beam direction. Experimental data: ●, this work; ▲, Kushawaha *et al.* (Ref. 6). Theoretical data: ○, Allan *et al.* (Ref. 7); ×, Fritsch (Ref. 15); — — —, Kimura *et al.* (Ref. 8). Note that in the latter calculation (Ref. 8) the anisotropy of the initial $3p$ state has not been specified. (b) ■, our experimental $\sigma(3s)$ and ●, $\sigma(3p)$ cross sections; cf. (a) and Fig. 3.

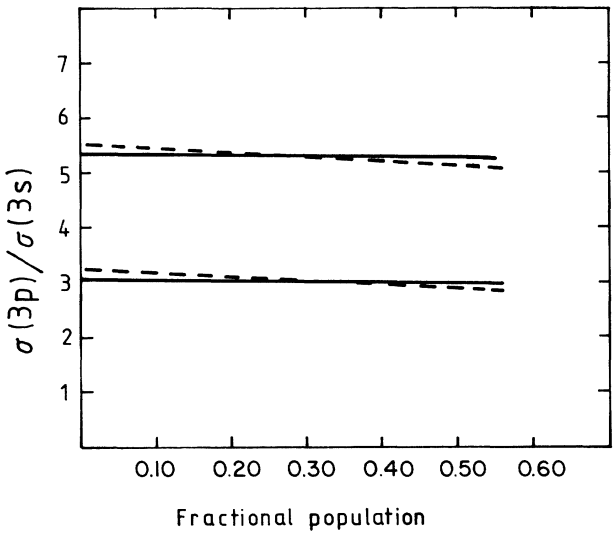

FIG. 5. $\sigma(3p)/\sigma(3s)$ as calculated for fractional substate populations different from those expected from an "ideal" optical pumping situation. Polarization vector (z direction) parallel (— — —) and perpendicular (——) to the projectile direction. Upper pair of curves, 0.5 keV proton energy; lower pair, 1 keV.

detailed initial substate populations [given, e.g., in Eq. (3)]. To demonstrate this, we have calculated $\sigma(3p)$ for a fractional $3p_z$ population of less than 0.56 (and accordingly higher fractional $3p_x$, $3p_y$ population) using the MO data of Allan *et al.*[7] (Fig. 5). Clearly, even a drastic variation will not change $\sigma(3p)$ much, irrespective of the light polarization direction; i.e., the Lyman-α production cross section is not very sensitive to the initial-state alignment as produced by linearly polarized light. This is due to the (accidental) interplay between the magnetic ($M_L=0,\pm1$) substate cross sections, and the mixture of hyperfine substates created by the pumping process.

IV. CONCLUSION

We have found a strong enhancement of Lyman-α production in collisions of H^+ with laser-excited Na($3p$) over Na($3s$) between 0.5 and 3 keV impact energy; this directly reflects the enhanced H($2p$) capture cross sections, in agreement with calculations by Allan *et al.*[7] and Fritsch.[15] The next steps to a more detailed understanding of the interaction process will have to include measurements differential with respect to the collision trajectory as well as the polarization properties of the emitted Lyman-α radiation. As in earlier experiments with (isotropic) ground-state atoms, one may now for the first time hope for a "complete experiment" with target atoms in initial states of controllable nontrivial shape and rotation.

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