Laser-Enhanced Lyman-α Production in Collisions between Hydrogen Ions and Sodium Atoms

V. S. Kushawaha, C. E. Burkhardt, and J. J. Leventhal
Department of Physics, University of Missouri-St. Louis, St. Louis, Missouri 63121

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The first measurement is reported of the absolute cross section as a function of kinetic energy for a process involving a state-selected excited reactant, namely near-resonant H^+-Na($3p$) charge exchange. The ion-beam experiments show enhanced production of $L\alpha$ from H^+-Na when the sodium is irradiated with laser light tuned to a D line. The results presented here demonstrate unambiguously that of H^+, H_2^+, and H_3^+, only H^+ leads to enhancement; however, the effect occurs only for energies below 30 eV.

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Laser-switched reactions, in which formation of a given product is enhanced by the presence of a laser beam, have been the subject of recent experimental[1-3] and theoretical[4,5] studies. Most of this work has centered on "laser-induced" processes for which the laser beam energy is tuned to that of a *virtual* state of the reactant species. However, a recent suggestion[6] that charge transfer involving one excited reactant could lead to the population inversion necessary for laser action at vacuum ultraviolet wavelengths has led to studies of laser-assisted reactions in which the laser is tuned to a *real* atomic transition.[3,7] In these experiments a pulse of moving hydrogen plasma was directed into a cloud of sodium vapor; illumination of the vapor with a laser tuned to a *D* line led to enhancement of the collision-produced Lyman-α ($L\alpha$) line. This enhancement was presumed to result from a larger cross section for the reaction

$$H^+ + Na(3p) \rightarrow H(n=2) + Na^+ \qquad (1)$$

than for the analogous H^+-Na($3s$) process. Since Reaction (1) is exothermic by 0.37 eV and the H^+-Na($3s$) process is endothermic by 1.74 eV, a larger cross section is expected for Reaction (1) because of the near resonance. In fact, gain at 1216 Å has recently been reported using the plasma-cloud technique.[8] In this Letter we report data which unequivocally show that Reaction (1) is responsible for the $L\alpha$ enhancement (and thus the observation of gain), while analogous H_2^+-Na and H_3^+-Na experiments show no enhancement at or near the $L\alpha$ wavelength. Furthermore, our measurements show that the enhancement in the H^+-Na experiments is a sensitive function of collision kinetic energy. We also report the absolute cross sections for $L\alpha$ production as a function of kinetic energy for H^+-Na($3p$) and H^+-Na($3s$) collisions. This is the first such measurement for a collision system involving a state-selected excited reactant. Absolute cross sections for H_2^+-Na and H_3^+-Na collisions are also presented.

Our experiments were performed by directing a mass-selected hydrogen-ion beam into a collision cell containing sodium vapor at $\sim 10^{13}$ cm^{-3}. Light from a single-frequency cw dye laser was directed into the collision cell and interpenetrated the ion beam; the laser power density was ~ 10 W/cm^2, which is sufficient to saturate the $3s \rightarrow 3p$ transition in sodium.[9] $L\alpha$ radiation was selected and detected, in a direction perpendicular to the beams, with a narrow band filter (maximum transmission at 1216 Å) and a solar-blind photomultiplier used in the counting mode. The laser beam was mechanically chopped and the photon signal detected in synchronization with the chopper, yielding "laser-on" and "laser-off" count rates designated A and B, respectively.

Figure 1 shows the enhancement factor, $\epsilon = (A-B)/B$, as a function of collision kinetic energy for H^+-Na collisions with the laser tuned both on and off the D_2 line. Although the graph extends only to 35 eV, measurements beyond this value show that $\epsilon = 0$ between 30 and 600 eV. This implies that the cross sections for production of $L\alpha$ in H^+-Na($3s$) and H^+-Na($3p$) collisions are identical in this energy range. The maximum value of ϵ is slightly less than 100%, which is consistent with the results of Dutta *et al.*[3]

By transmitting either H_2^+ or H_3^+ with the mass spectrometer it was possible to obtain similar data for H_2^+-Na and H_3^+-Na collisions. In these cases detection of photons at the $L\alpha$ wavelength is not necessarily indicative of charge transfer or even of $L\alpha$ radiation from a hydrogen atom. Other processes, such as dissociation, dissociative charge transfer, and simple excitation, are possible so that, because some of the bands from the Lyman and Werner systems of molecular hy-

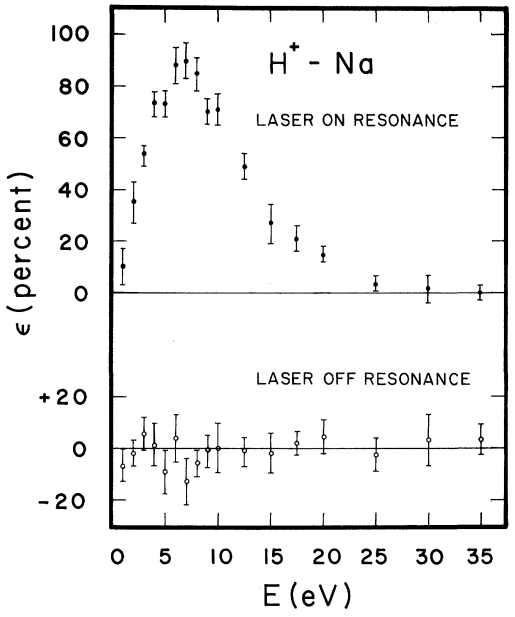

FIG. 1. The enhancement factor, ϵ, as a function of collision kinetic energy, E, taken with the laser tuned to the D_2 line and slightly detuned from this line. The error bars represent statistical uncertainty.

drogen radiate within the bandpass of the $L\alpha$ filter, a variety of processes may be detected simultaneously. Nevertheless, experiments were performed for H_2^+-Na and H_3^+-Na collisions to determine whether these ions play any role in enhancing the $L\alpha$ signal in the plasma experiments. Data analogous to those displayed in Fig. 1 show that no enhancement occurs for either collision system over the range 1–600 eV. This observation unambiguously establishes Reaction (1) as the process responsible for the enhancement in the hydrogen plasma experiments.

In addition to the direct determinations of ϵ discussed above, absolute cross sections for relevant processes were measured as functions of collision energy. This was accomplished by first calibrating the detection system against measured cross sections for $L\alpha$ emission in He^+-H_2 collisions,[10,11] and determining the Na atom density by use of a previously described technique which relies on both surface ionization and ion impact.[9] Figure 2 shows H^+-Na cross sections for both reactant Na($3s$) and Na($3p$); those for H^+-Na($3p$) were obtained by combining our enhancement-factor data with our measurements of the H^+-Na($3s$) cross section. Because of difficulties associated with precise measurement of the primary ion current at the lowest kinetic energies, absolute cross sections are not reported below 5 eV. It should be noted, however, that in the measurements of ϵ the primary ion current cancels, so that the data displayed in Fig. 1 are considered to be reliable down to 1 eV. The energy dependence of the H^+-Na($3s$) cross section is typical of endothermic processes, and appears to extrapolate to the proper threshold. As discussed above, the cross section for the slightly exothermic H^+-

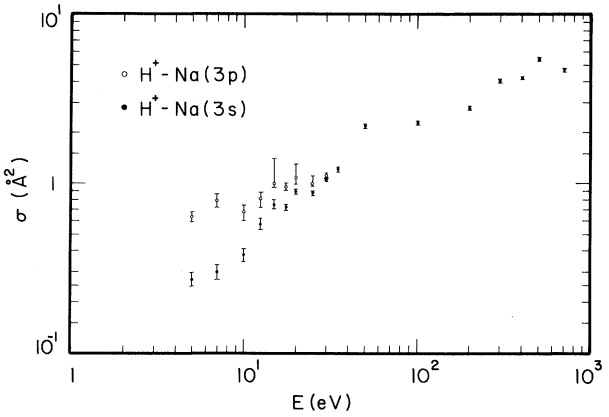

FIG. 2. Cross sections, σ, as a function of kinetic energy, E, for H^+-Na($3s$) and H^+-Na($3p$) collisions. The error bars represent statistical uncertainty.

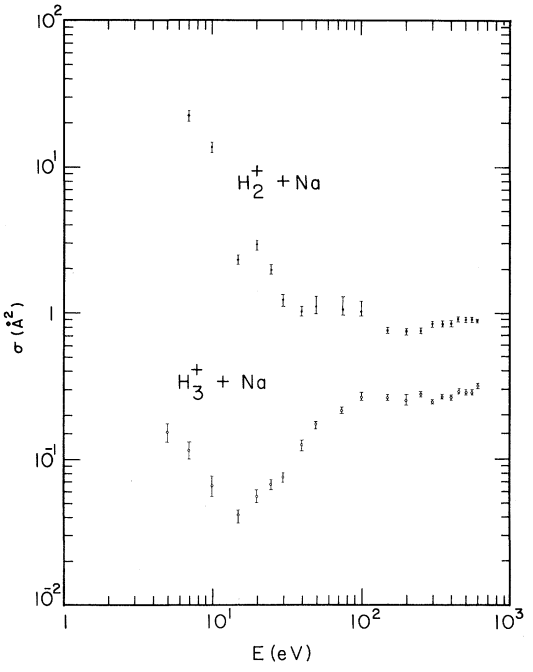

FIG. 3. Cross sections, σ, for H_2^+-Na and H_3^+-Na collisions as described in the text. The error bars represent statistical uncertainty.

Na($3p$) charge exchange differs from the H^+-Na($3s$) values only below 30 eV where it decreases slowly with decreasing energy. This behavior is in qualitative agreement with the well-known theoretical treatment of nonresonant charge transfer of Rapp and Francis.[12]

Finally, for completeness, we display in Fig. 3 the absolute cross sections for H_2^+-Na and H_3^+-Na collisions. Recall that because no enhancement was observed at any kinetic energy for either of these systems the cross sections are the same for both Na($3s$) and Na($3p$). Of course in these molecular systems the additional vibrational and rotational states available to reactants and products make it difficult to say anything about *specific* reaction channels. As discussed above, detection of radiation having wavelengths within the bandpass of the filter could be indicative of the occurrence of any combination of the variety of processes possible in these molecular systems. The increasing cross section with decreasing kinetic energy in both the H_2^+ and H_3^+ cases suggests that one or more of the possible resonant (thermoneutral) processes is occurring. The structure for H_3^+-Na is evidence that additional reaction channels play a role at the higher kinetic energies.

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