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Observation of Structure in Laser-Induced Penning and Associative Ionization in Crossed-Beam Na + Na Collisions

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The results of double-laser experiments in which Na_2^+ and Na^+ are produced in crossed alkali beams under single-collision conditions in the presence of strong optical fields are reported. Structure in the mass-selected product ion intensity as a function laser frequency is observed when the optical field is strongly focused and tuned far off atomic or dimer transitions. These measurements are the first to show that the nuclear motion of the quasimolecular collision intermediate plays an important role in laser-induced collisional ionization.

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Laser-switched or laser-modified inelastic and reactive collisions are presently the object of intensive theoretical[1-3] and experimental study[4-7] because they offer the possibility of controlling both the relative and absolute probability of competing exit channels. The influence of the laser field is to modify the *electronic* states during the course of a collisional encounter. It is even possible to alter the outcome of a collision without a net transfer of energy between the atomic system and the radiation field.[8] In all cases "laser-induced" collisions are characterized by atomic field interactions which are nonresonant with respect to dipole-allowed transitions of the separated collision partners. It is important to distinguish these processes from multiphoton infrared absorption experiments in which nuclear motion absorbs photon energy directly. To date, observation of laser-induced energy transfer,[4,6] charge transfer,[5] and collisional ionization[7] have been established. We discuss here new results on Penning and associative ionization. With the optical-field intensity increased by over two orders of magnitude, we observe an entirely new domain of rich structural features. The observed collisions are

$$Na + Na \xrightarrow{\hbar\omega} Na_2^+ + e \quad \text{(associative ionization)},$$

$$Na + Na \xrightarrow{\hbar\omega} Na^+ + Na + e \quad \text{(Penning ionization)},$$

where the symbol $\hbar\omega$ indicates "in the presence of the laser field".

The crossed-beam apparatus is a high-vacuum cylindrical stainless-steel chamber in which two alkali atomic beam sources are mounted at right angles in a horizontal plane. Two laser beams enter from opposite ports, in the same plane and all four beams overlap in the interaction region at the chamber center. Kingston-Langmuir hot-wire detectors monitor effusive flux from the

alkali ovens; a quadrupole mass filter unit, mounted vertically above the intersection plane, detects the product ions. Resolution of the quadrupole mass filter is $\simeq 100$ which is more than sufficient to clearly separate Na^+ from Na_2^+. A copper baffle in thermal contact with a liquid-nitrogen resevoir completely surrounds the interaction and detection region. The atomic beam sources are of a double-oven design with the exit orifice of each separately heated from the main body. The orifices are heated ~200 °C above the main ovens to prevent clogging and suppress dimer contamination. Both atomic beams are collimated before entering the interaction region. Light sources are flash-lamp-pumped tunable dye lasers synchronized together and with a box car integrator/amplifier used to record the particle multiplier signal. The output of the box car is recorded as a function of laser frequency on a strip chart recorder. Typical operating conditions are (a) sodium-atom density at collision region $\simeq 3\times10^9$ atoms cm^{-3}, (b) focused laser intensity $\simeq 3\times10^7$ W/cm^2, (c) mass-analyzer collection efficiency $\lesssim 10\%$, and (d) particle-multiplier gain $\lesssim 10^8$. From these conditions, the geometry of the interaction volume, and the magnitude of the final output signal (~10^{-12} C per laser pulse) we estimate a cross section of ~10^{-17} cm^2 for those laser-induced processes. This result is in rough agreement with theoretical prediction.[9]

The simplest experiment is to leave laser 2 turned off and laser 1 unfocused ($I \simeq 5\times10^4$ W/cm^2). Under these conditions only two peaks in the laser scan appear, corresponding to atomic resonance transitions, $Na(^2S_{1/2}) \rightarrow Na^*(^2P_{3/2}, ^2P_{1/2})$. Laser power saturates both transitions. The same very simple spectrum was first reported by Lucatorto and McIlrath[10] and is obtained both for Na^+ and Na_2^+ production. As discussed elsewhere,[11] from these spectra and measurements of ion intensity versus laser intensity we interpret this result as a two-step process in which population of $Na^*(^2P_{3/2}, ^2P_{1/2})$ is followed by associative ionization, i.e.,

$$Na(3s) + Na(3s) + 2\hbar\omega \rightarrow Na^*(3p) + Na^*(3p)$$

followed by

$$Na^*(3p) + Na^*(3p) \rightarrow Na_2^+ + e.$$

The appearance of atomic ions may be due to either photodissociation of Na_2^+, or a direct, laser-induced Penning ionization. The potential curves for Na_2 and for Na_2^+ in Fig. 1 indicate that

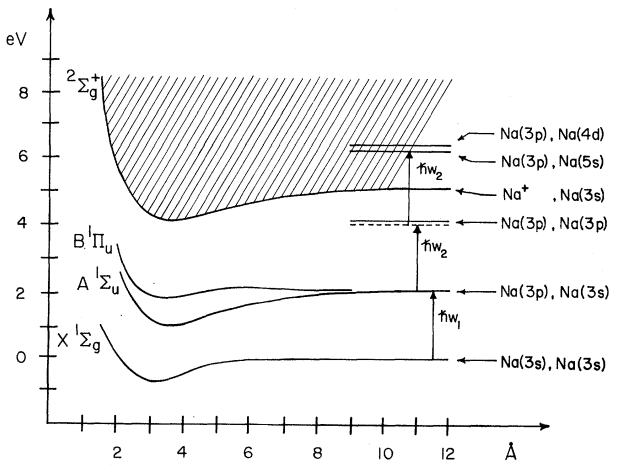

FIG. 1. Potential energy curves of Na_2 and Na_2^+ for well-known singlet levels. Curves taken from data in references 12, 13, 17, and 18.

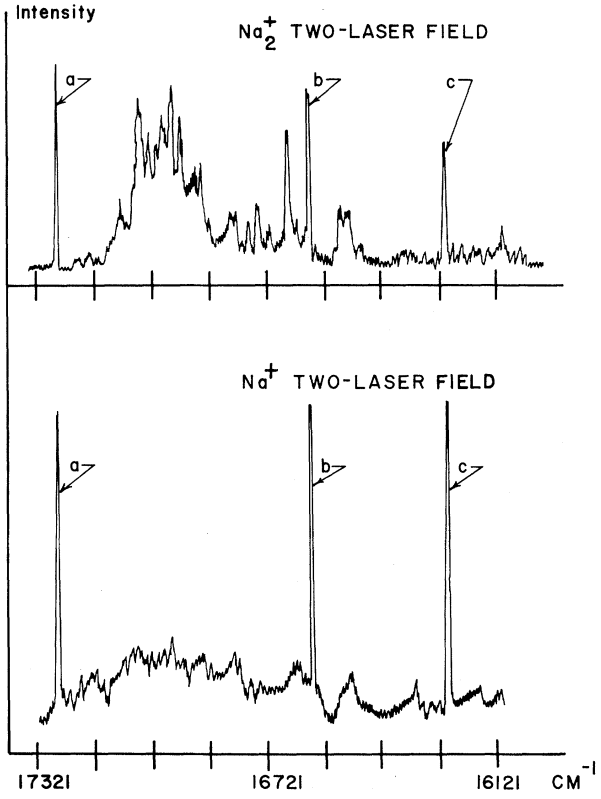

FIG. 2. Spectra of Na_2^+ and Na^+ as function of focused-laser frequency. The zero-intensity baseline with both lasers off is as shown at the extreme left of each spectrum.

molecular curves constructed from $Na^*(3p)+Na^*(3p)$ atomic levels are very probably degenerate with Na_2^+ near the minimum of the ion dimer potential. However, we are not aware of any calculations of these excited molecular potentials. We emphasize that no ion signal is observed when the laser is tuned off the atomic resonance lines.

In contrast Fig. 2 shows the spectrum obtained from a two-laser experiment in which laser 1 remains unfocused and tuned to the $(^2P_{3/2})$ atomic resonance line. Laser 2 is focused to $\simeq 3\times10^7$ W/cm^2 while scanning over a range of frequencies. Two narrow prominent peaks in the Na_2^+ spectrum (labeled *a* and *b* in Fig. 2) are the results of processes involving two-photon atomic transitions. Peak *a* arises from

$$Na(3s^2S_{1/2})+2\hbar\omega \rightarrow Na^*(4d^2D_{5/2,3/2}),$$

followed by associative ionization

$$Na^*(4d)+Na^*(3p) \rightarrow Na_2^+ + e. \quad (1)$$

The corresponding peak in the Na^+ signal is due to resonance-enhanced three-photon photoionization. Similar remarks apply to peak *b* in which a two-photon transition populates $Na^*(5s^2S_{1/2})$. Peak *c* arises from two, sequential single-photon transitions:

$$Na(3s^2S_{1/2})+\hbar\omega \rightarrow Na^*(3p^2P_{3/2}) \quad \text{(laser 1)} \quad (2)$$

and

$$Na^*(3p^2P_{3/2})+\hbar\omega_2 \rightarrow Na^*(5s^2S_{1/2}) \quad \text{(laser 2)}, \quad (3)$$

again followed by associative ionization.

In marked contrast the off-resonance signal is weaker and structural features are markedly broader than the three principal peaks. On the long-wavelength side, onset of Na_2^+ begins around 16 000 cm^{-1}, reaches a very broad maximum about 16 940 cm^{-1}, and declines rapidly after passing through peak *a* at 17 274 cm^{-1}. Regularly spaced secondary maxima with approximate spacings and widths of about 120 cm^{-1} and 20 cm^{-1}, respectively, can be discerned. We interpret these off-resonance effects as laser-induced associative ionization. This process is

$$Na^*(3p)+Na(3s)+\hbar\omega_2 \rightarrow Na_2^+ + e, \quad (4)$$

where $\hbar\omega$ is not resonant with the atomic transitions of the separated partners. The laser-induced process may be regarded as preparation of a virtual state (dashed line in Fig. 1) by the strong laser field immediately followed by an associative ionization collision or as the absorption of radiation by a short-lived quasimolecular collision intermediate. From either viewpoint, inspection of Fig. 1 indicates that onset should occur when $\hbar\omega_2$ satisfies the energy defect of the process, $\simeq 2.0$ eV (16 131 cm^{-1}). The observed onset is in agreement with this energy requirement. As a check on the energy threshold, measurements were extended to the red as far as 15 625 cm^{-1}. No additional Na_2^+ signal was observed. We interpret the secondary peaks as population of vibrational states in Na_2^+. The peak spacings are consistent with vibrational energy levels separations calculated by Bardsley, Junker, and Norcross,[12] and Dalgarno, Cerjan, and Docker.[13] Peak widths are attributed to lifetime broadening of the quasimolecular collision intermediate which is estimated to be $\simeq 20$ cm^{-1}.

Because of a large photodissociation cross section[14] for Na_2^+, off-resonant Na^+ production is probably due to this process. However, a direct laser-induced Penning process cannot be ruled out, and the two possibilities cannot be distinguished in the present experiment.

Several checks were carried out to verify that dimers in the beams were not responsible for the observed ion signals. First the temperature of the oven nozzles was maintained at about 800 °K. A simple thermodynamic calculation shows that the dimer concentration at this temperature is 0.032% of the monomer. Therefore any two-body collisional process involving dimers would be required to have a cross section $\simeq 10^{-12}$ cm^2 to agree with observed signal strength. The experiment was repeated with only a single alkali beam. No Na_2^+ signal clearly distinguishable from background was observed. The atomic ion was observed only at resonance peaks *a* and *b*, proving that atomic photoionization dominates at these frequencies. Failure to detect a signal from Na_2^+ implies an upper limit of $\simeq 0.1\%$ on dimer ion concentration in the effusive alkali beam and proves that photoionization of dimers is not an artifact of the experiment. Further, our observed dimer ion spectra do not match those from multiphoton ionization of neutral dimers formed in molecular jets.[15-16]

These results clearly show that laser-induced inelastic processes cannot be described uniquely by simple electrostatic long-range interactions. The rich structural features in laser-induced collisional ionization cross sections indicate that vibrational and perhaps rotational motion of the quasimolecular intermediate plays an important role. It is hoped that these observations will pro-

vide fresh stimulus for theoretical development. Experimental work is continuing on alkali and rare-gas systems.

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Major Disruptions in the TOSCA Tokamak

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The major disruption has been studied in detail on the TOSCA tokamak with the use of approximate helical coils. The $m=3$, $n=2$ and $m=5$, $n=3$ modes have been clearly observed for the first time before a major disruption. The amplitude of the $m=2$, $n=1$ mode at the rational surface is about 4% and the $m=3$, $n=2$ mode amplitude ~4.5% before a major disruption. When the $m=2$, $n=1$ and $m=3$, $n=2$ approximate helical coils are energized with large coil currents, a major disruption occurs.

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Further improvements of plasma parameters in present-day tokamak devices are limited by the major disruption.[1] In the next generation of tokamaks, the major disruption can be a severe problem because large forces and voltages are generated by the sudden decrease in plasma current following the disruption. An understanding and a means of controlling this dangerous instability is therefore very important for the future of the tokamak device.

TOSCA is a small air-core tokamak, which can produce noncircular plasmas.[2] The major radius of the vacuum vessel is 30 cm and the minor radius 10 cm. Central electron temperatures of 300 eV and central electron densities of 3×10^{13} cm^{-3} are routinely obtained, with a plasma current of 12 kA, a loop voltage of 2.5 V and a safety factor at the edge of the plasma, q_a, of 3.5.

A series of large saddle coils were used to detect the helical radial field perturbations produced

