Observation of laser-induced associative ionization in crossed-beam Na+Li collisions

P. Polak-Dingels, J. Keller, and J. Weiner
Department of Chemistry, University of Maryland, College Park, Maryland 20742

J. -C. Gauthier
Institut d'Electronique Fondamentale, Bâtiment 220, Université de Paris-Sud, 91405 Orsay, France

N. Bras
Laboratoire de Spectroscopie Atomique, Université Pierre et Marie Curie, 4, Place Jussieu 75230 Paris, France
(Received 17 February 1981)

We report the observation of $NaLi^+$ produced by crossed-beam collisions of Na and Li in the presence of a single laser field. The magnitude of the cross section and its dependence on laser intensity are in accord with a simple second-order perturbation theory.

During the past three years a growing body of experimental evidence[1-7] has demonstrated that collisional dynamics may be dramatically altered in the presence of intense laser radiation. In these phenomena the light fields play an active role in the overall interaction and are not simply the means by which initial and final states are prepared or detected.

In previous papers[3,4] we reported laser-induced production of Li_2^+ and Na_2^+ in collisions between identical particles. We report here the observation of $NaLi^+$ when separate beams of Na and Li atoms are crossed in the presence of a laser field. The collisional equation is

$$Na^*(3p) + Li(2s) + \hbar\omega \rightarrow NaLi^+ + e\,. \qquad (1)$$

This new observation differs from previous reports in two important ways. First, the associative process is heteronuclear. It demonstrates that a laser field can induce bonding between chemically distinct particles in a two-body collision. Second, the effect is simpler to analyze because

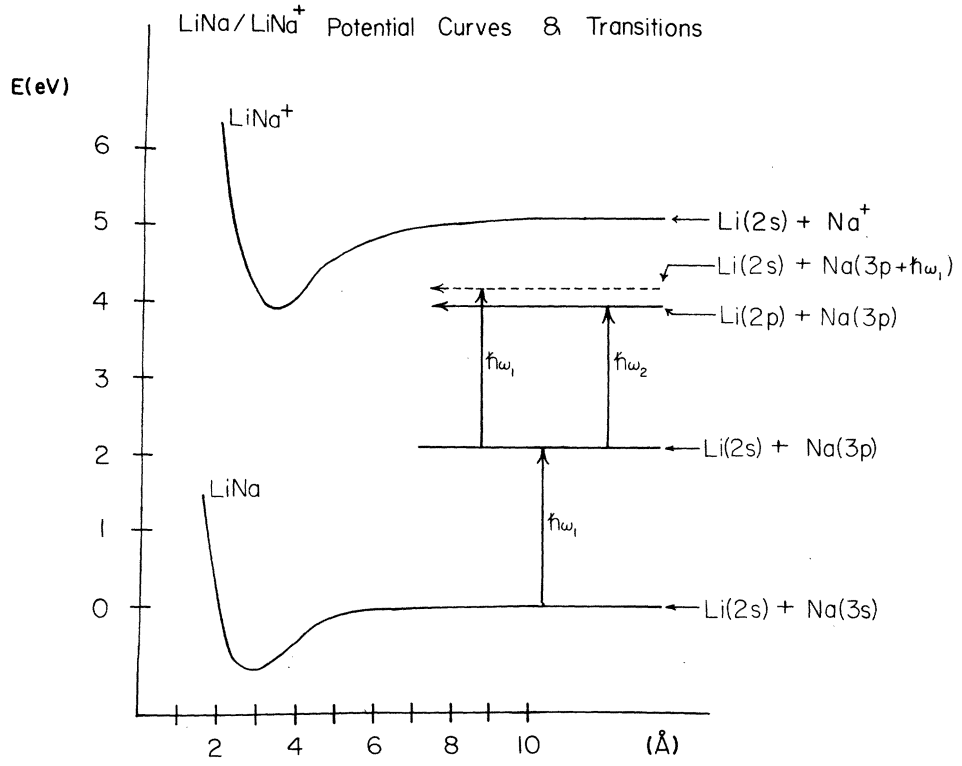

FIG. 1. Potential-energy levels of the Na, Li system.

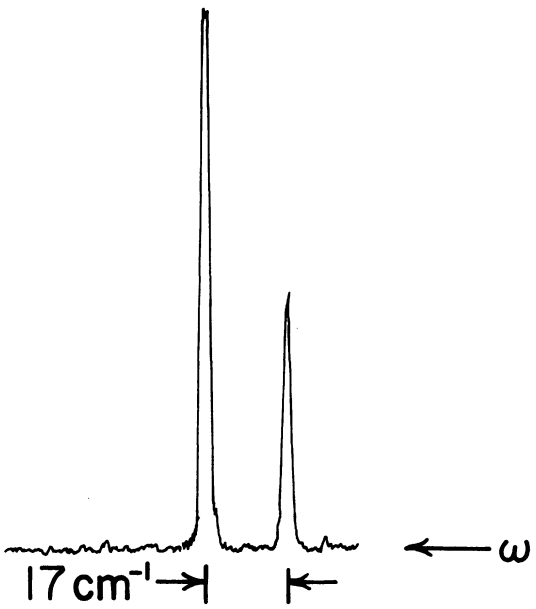

FIG. 2. Mass spectrum of $NaLi^+$ as a function of laser frequency. Peak positions are within 1 cm^{-1} of $Na(^2P_{3/2}, {}^2P_{1/2})$ transitions.

only one of the partners (sodium) interacts strongly with the light field. In previous experiments field excitation of colliding partners was necessarily symmetrical.

The experimental apparatus is quite similar to that described in earlier reports.[4,5] The lithium atomic-beam source was designed as a "double oven" similar to the sodium source but modified to permit continuous operation at temperatures near 1000 °C. The atomic beams were maintained at right angles in the horizontal scattering plane but moved radially somewhat closer together so as to increase particle-number density in the collision zone. Typical operating conditions were (a) alkali atom density $\simeq 10^{12}$ atoms cm^{-3}, (b) unfocused laser intensity $\simeq 5\times10^4$ W cm^{-2}. Invoking the usual assumptions of instrumental detection sensitivity and efficiency,[4] we estimate the cross section for the laser-induced processes $\sim 10^{-19}$ cm^2 which is in agreement with a perturbation calculation of collisional ionization.[8]

Figure 1 depicts the relevant potential curves and the optical-collisional interaction sequence. In the first step the laser field tuned to the $Na(3s \rightarrow 3p)$ transition excites a *virtual* state of sodium [approximately midway between $Na^*(5s)$ and $Na^*(4d)$ levels] at *twice* the resonant frequency. The second step, associative ionization, then takes place between the "dressed" sodium atom and ground-state lithium.

Figure 2 shows two peaks in the $NaLi^+$ spectrum at laser frequencies within 1 cm^{-1} of the $Na^*(^2P_{3/2}, {}^2P_{1/2})$ levels. No other $NaLi^+$ peaks were observed.

Theory[8,9] predicts that in the weak-field limit, intensity of the $NaLi^+$ should be linear with laser intensity. Figure 3 shows the results of experiments in which the laser beam was systematically

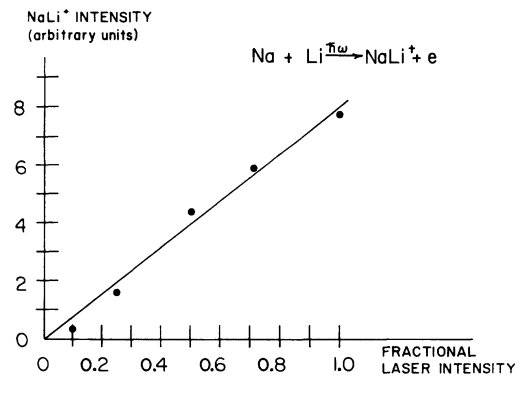

FIG. 3. Ion intensity vs laser intensity for laser-induced process. Unattenuated laser peak power $\simeq 5\times10^4$ W cm^2.

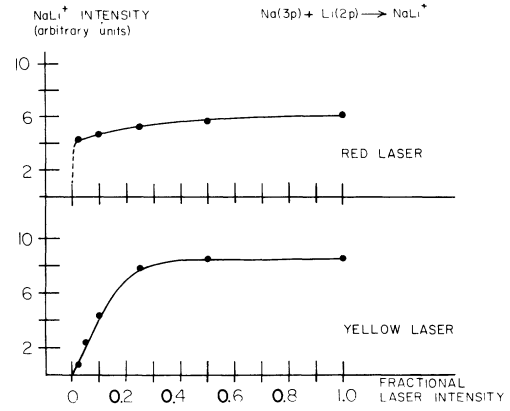

FIG. 4. Ion intensity vs laser intensity for two-laser experiment in which each laser resonantly populates an excited atomic state.

attenuated by a set of calibrated neutral density filters. Linearity of the plot is self-evident. This result contrasts sharply with those obtained from conventional associative ionization between excited states of Na and Li. In a separate set of experiments with two lasers, tuned,respectively, to the Na*($3p$) and Li*($2p$) transitions, we also observe $NaLi^+$ production. In this case, however, we speculate that the process may proceed via a two-step mechanism in which the initial step is an inverse predissociation,

$$Na(3p) + Li(2p) \rightarrow NaLi^*, \tag{2a}$$

followed by photoionization of the quasimolecule,

$$NaLi^* + h\omega \rightarrow NaLi^+ + e. \tag{2b}$$

Saturation of $NaLi^+$ intensity at high laser power is a necessary consequence of such a two-step mechanism.[10] Attenuation experiments on each laser were repeated with the result presented in Fig. 4. The saturation characteristic is clearly evident.

It is interesting to speculate on two-laser experiments in which the first laser is tuned to the Na($3s \rightarrow 3p$) frequency but the second is nonresonant with respect to any atomic transition. If the first Na($3s \rightarrow 3p$) transition is "uncorrelated" with the collisional interaction,[6] then we may expect enhancement of the ion signal by a second nonresonant laser, the light field of which has no definite phase relationship with the first. The results of the other two-laser experiments, both resonant and nonresonant, will be reported shortly.

Support from the Office of Naval Research under Contract No. ONR N0014-78C-0823 and from the National Science Foundation under Grant No. NSF INT 79-21253 is gratefully acknowledged.

[1]R. W. Falcone, W. R. Green, J. C. White, J. F. Young, and S. E. Harris, Phys. Rev. A 15, 1333 (1977).
[2]W. R. Green, J. Lukasik, J. R. Willison, M. D. Wright, J. F. Young, and S. E. Harris, Phys. Rev. Lett. 42, 970 (1979).
[3]A. V. Hellfeld, J. Caddick, and J. Weiner, Phys. Rev. Lett. 40, 1369 (1978).
[4]P. Polak-Dingels, J.-F. Delpech, and J. Weiner, Phys. Rev. Lett. 44, 1663 (1980).
[5]P. Polak-Dingels and J. Weiner, J. Chem. Phys. 74, 508 (1981).
[6]P. Thomann, K. Burnett, and J. Cooper, Phys. Rev. Lett. 45, 1325 (1980).
[7]P. Hering, P. R. Brooks, R. F. Curl, R. S. Judson, and R. S. Lowe, Phys. Rev. Lett. 44, 687 (1980).
[8]J. Weiner, J. Chem. Phys. 72, 2856 (1980).
[9]S. E. Harris, unpublished lecture notes available from Stanford University, Department of Electrical Engineering, Palo Alto, California 94305.
[10]The model-potential calculation of the $NaLi^+$ binding energy by P. Habitz and W. H. E. Schwartz [Chem. Phys. Lett. 34, 248 (1975)] implies that the overall process (2) is endoergic by 0.2 eV, and therefore *conventional* associative ionization cannot explain the results. An independent *ab initio* calculation of the ground $^2\Sigma^+$ state of $NaLi^+$ by W. Stevens of the National Bureau of Standards confirms this conclusion.