Simultaneous Laser- and Ion-Beam Excitation of a Na-Beam Target

D. Schneider, V. Pfeufer,[a] W. Stöffler,[b] R. Bruch,[c] H. G. Berry, L. P. Somerville, J. E. Hardis,[d] P. Arcuni,[d] P. Seidel,[e] and C. F. Moore[e]
Argonne National Laboratory, Argonne, Illinois 60439
(Received 22 June 1983)

We report the first measurement of Na and Na^+ autoionization spectra following collisions of 1-MeV He^+ ions on selectively excited Na $3p\ ^2P_{3/2}$ target atoms. The atomic states were populated with a cw dye laser which was tuned to the Na $3s\ ^2S_{1/2} \rightarrow$ Na $3p\ ^2P_{3/2}$ transition. The simultaneous laser- ion-beam excitation technique produces significant intensity variations particularly for the decay of the $Na^+\ 2s2p^63s\ ^{1,3}S$ and the $Na^+\ 2s2p^63p\ ^{1,3}P$ states. The technique may provide additional information for line identification, excitation mechanisms, and angular distribution measurements.

PACS numbers: 34.50.Hc, 32.80.Dz

The high-intensity, narrow spectral bandwidth, and tunability of dye lasers together with fast-ion–atom collisions allow state-selective excitation plus ionization of atoms: The laser excites specific outer-shell states of an atom, which subsequently interacts with a fast-ion beam. This two-step excitation process may enhance the creation of short-lived autoionizing or Auger states which are not efficiently created in pure ion-atom collision processes. In addition, the high degree of polarization of laser beams may predominantly populate specific magnetic substates of the outer-shell target states.

Electron scattering experiments with laser-excited atoms have been performed by Hermann and Hertel.[1] Measurements of differential cross sections for laser-excited Na atoms at thermal collision energies are described by Düren.[2] The technique of uv absorption spectroscopy of laser-pumped vapors has been developed by McIlrath and co-workers.[3] The first successful experiment using a cw dye laser tuned to the resonance lines of Na and Ba and of synchroton radiation to observe electrons produced by photoionization has been performed quite recently by Wuilleumier *et al.*[4,5] In this work the combined laser-excitation, ion-atom collision method has been used to study the Na L autoionization spectrum.

The experimental arrangement is shown in Fig. 1.

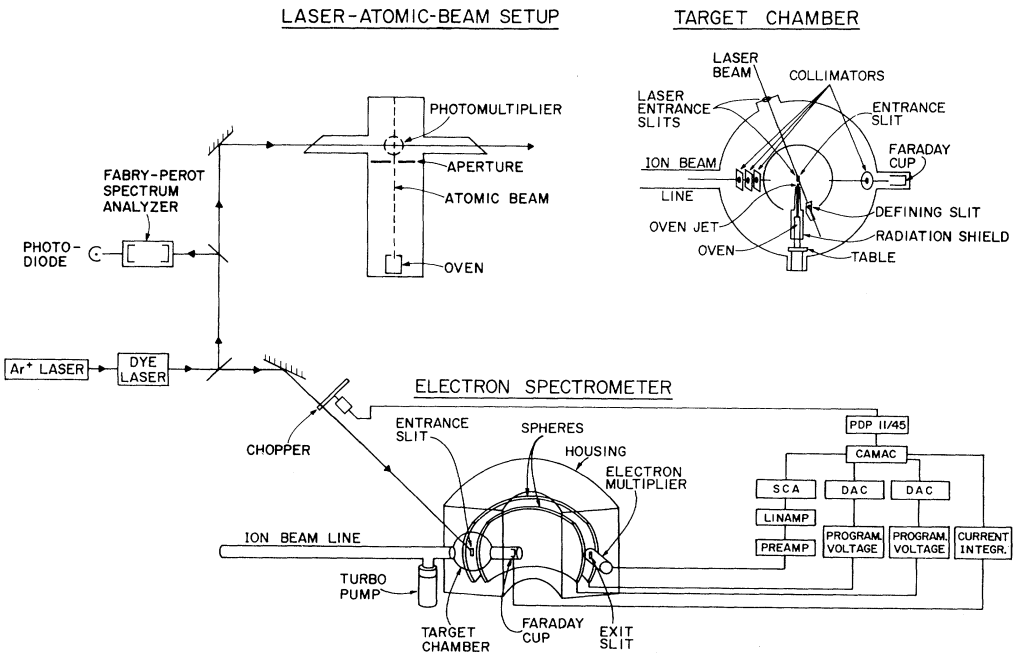

FIG. 1. Experimental arrangement.

An argon-ion-laser–pumped single-mode cw dye ring laser (Spectra Physics model 380 D) was tuned to the Na $3s\,^2S_{1/2} \rightarrow 3p\,^2P_{3/2}$ transition. The laser linewidth was about 1 MHz and the output power up to 600 mW. To control the laser wavelength a small part of the laser light was sent into a laser–atomic-beam apparatus. The laser light intersected orthogonally a collimated atomic beam of natural sodium, produced with a resistance heated tantalum oven. The collimation of the atomic beam reduced the Doppler width in the direction of the incident light to less than 30 MHz. The laser was tuned to the hyperfine transition with total angular momenta F equal to 2 and 3 for the lower and upper state, respectively. The resonance fluorescence was detected by a photomultiplier in a direction perpendicular to both the atomic and the laser beam.

In the scattering chamber natural sodium was heated in a magnetically shielded resistance heated oven approximately at 300 °C and effused through a 1 mm collimating aperture. The target density times thickness was about 3×10^{-3} Torr·mm. The laser beam intersected the scattering volume at an angle of 50° relative to the atomic beam axis. A collimated 1 MeV He^+ beam crossed the Na target jet at right angles. In order to detect the autoionization electrons of the laser- ion-beam excited atoms the oven was mounted in front of a spherical plate McPherson electron analyzer, which has been described in detail by Matthews. The secondary electron emission was observed at 90° with respect to the incident He^+ and Na beam axis. The laser intensity was chopped with a frequency of about 170 Hz, and autoionization spectra in the energy region from 16 to 33 eV were taken both with and without additional laser excitation.

A high-resolution Na L autoionization spectrum (100 meV at 25 eV) as produced with 1 MeV He^+ on Na is plotted in three parts in Fig. 2. The main peaks are numbered 1 to 8 in order of increasing energy and the spectral assignments are given in Table I. Calibration of the energy scale was achieved by using the

$$Na^+\ 2s2p^6 3s\ ^3S_1 \rightarrow 2s^2 2p^5\ ^2P^0_{3/2}$$

Coster-Kronig transition.[7,10]

The effects of additional laser excitation are deduced from comparison with difference spectra of laser- ion-beam and pure ion-beam produced spectra (chopped laser beam) in Fig. 2. The total Na L autoionization spectrum measured in three parts and the corresponding difference spectrum are plotted in Figs. 2(a)–2(c). A dip in the difference spectrum indicates an enhancement in the correspond-

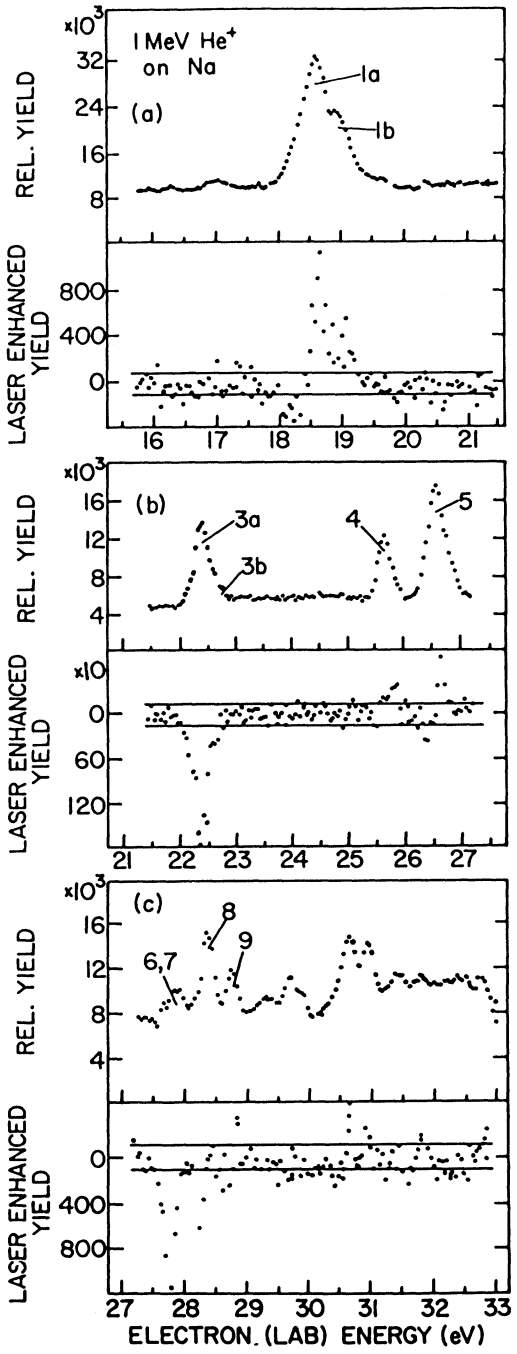

FIG. 2. Na L autoionization spectrum (15 to 33 eV) following 1 MeV He^+ impact on Na beam target; line assignments are given in Table I. Observation angle is 90° with respect to the beam axis; the energy resolution is 100 meV at 25 eV. The upper spectra in (a)–(c) are the autoionization spectra without laser excitation; the lower spectra are the difference spectra obtained from the measurements without and with $3s\,^2S_{1/2}$–$3p\,^2P_{3/2}$ laser excitation of Na. The solid lines indicate the standard deviation.

TABLE I. Spectroscopic assignment of lines observed in a Na autoionization spectrum produced by 1 MeV He^+ ions (Fig. 2).

Peak number	Peak energy (eV)	Spectroscopic assignment initial state	final ionic state	Transition energy (eV)
		$Na^+(2s2p^63s)^{3,1}S$	$(2s^22p^5)^2Po_{1/2,3/2}$	
1a	18.6	$^3S_1$	$^2P_{1/2}$	18.48[a]
		$^3S_1$	$^2P^0_{3/2}$	18.65[a]
1b	19.1	$^1S^0$	$^2P^0_{1/2}$	18.98[a]
		$^1S^0$	$^2P^0_{3/2}$	19.15[a]
2				
		$Na^+(2s2p^63p)^{1,3}P^0$	$(2s^22p^5)^2P^0_{1/2,3/2}$	
3a	22.4	$^3P^0_{1/2}$	$^2P^0_{1/2}$	22.32[a]
		$^3P^0_{1/2}$	$^2P^0_{3/2}$	22.49[a]
3b	22.6	$^1P^0_1$	$^2P^0_{1/2}$	22.49[b]
		$^1P^0_1$	$^2P^0_{3/2}$	22.66[b]
		$Na(2s\ 2p^63s3p)$	$(2p^53p)$	24.4[a]
		$Na(2s^22p^53s^2)^2P^0$	$(2p^6)^1S$	
4	25.6	$^2P^0_{3/2}$	^1S	25.63[a]
		$^2P^0_{1/2}$	^1S	25.79[a]
		$Na^+(2s2p^64s)^{1,3}S$	$(2s^22p^5)^2P^0_{1/2,3/2}$	
5	26.4	$^3S_1$	$^2P^0_{1/2,3/2}$	26.46[c]
		$^1S_0$	$^2P^0_{1/2,3/2}$	26.64[c]
6,7	27.5	$Na^+(2s2p^64p)^{1,3}P^0$	$(2s^22p^5)^2P^0_{1/2,3/2}$	
		$^1P^0_1$	$^2P^0_{1/2}$	27.73[b]
	27.8	$^1P^0_1$	$^2P^0_{3/2}$	27.89[b]
8	28.3	$Na(2s^22p^53s^3p)$	$(2s^22p^6)^1S$	28.24–38.37[a]
	33	$Na^+(2s2p^6nl)$ Series Limit	$(2s^22p^5)^2P^0_{1/2,3/2}$	32.8

[a]Breuckmann *et al.* (Ref. 7).
[b]Lucatorto *et al.* (Ref. 8) and Sugar *et al.* (Ref. 9).
[c]Ross *et al.* (Ref. 10) and H. W. Wolff *et al.* (Ref. 11).

ing line due to additional laser excitation. It is evident from these spectra that the most drastic intensity variations are observed for the lines labeled $1a$, $1b$ and $3a$, $3b$ and for lines observed between 27 and 29 eV. Minor variations which are exceeding the limits of the standard deviations as indicated in the spectra in Fig. 2 are further observed around 26 and 31 eV.

Lines $1a$ and $1b$ at 18.65 eV are due to the $2s$ ionization and the following Coster-Kronig transition:

$$2s2p^63s\ {}^1S_0,\ {}^3S_1 \rightarrow 2s^22p^5\ {}^2P_{1/2},\ {}^2P_{3/2} + \epsilon p.$$

The observed peak in the difference spectrum means a decrease of the spectral line intensity if additional laser excitation is applied. This results from a decrease of the population of the $2s2p^63s$ initial configuration due to the laser excitation of the $3s$ electron.

Lines $3a$ and $3b$ at 22.4 eV are due to $3s$ ionization and simultaneous excitation of a $2s$ electron to the initial states $2s2p^63p\ {}^{1,3}P^{\circ}$. It should be noted that this line group is strongly excited in the 1 MeV He^+ on Na collision (Refs. 8 and 12). Lines $3a$ and $3b$ are found to be most affected by the simultaneous laser- ion-beam excitation due to an increased probability for the population of the $2s2p^63p$ configuration.

Lines 6, 7, and 8 are due to the decay of the initial configurations as $2s2p^63s3p$ and $2s2p^64p$ which can be produced by quadrupole excitation of $2s$ and $2p$ electrons.[7-9] Again the laser excitation changes the population of the initial configuration and enhances therefore the probability for simultaneous $3s$ ionization and $2s \rightarrow nl$ excitation in the collision.

Less significant intensity variations around 26 and 31 eV could be explained in a similar manner. A detailed spectroscopic analysis will be given in a forthcoming paper. The summed intensities in the peaks and "dips" as observed in the difference spectra reflect the rearranged population of configurations with $3s$ and $3p$ electrons in case of additional laser excitation. The greater sharpness of the difference spectra may originate from different multiplet populations produced in the collision with and without laser excitation.

The laser-excitation collision technique for the investigation of Auger and autoionization spectra has been used for the first time. Intensity changes of the laser- ion-beam excited spectra compared to pure ion-beam excitation have been observed. Our measurements demonstrate that selective laser excitation may be used to obtain more detailed information for line identification and excitation mechanisms from Auger and autoionization spectra compared with normal optical or ion-beam techniques. This new technique may open the possibility to study the electron angular distribution as a function of the initial magnetic substate population of the target atoms. As a result of the lack of theoretical model calculations for energetic laser assisted ion-atom collision processes the conclusions are limited at present.

This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. W-31-109-Eng-38 and the Robert A. Welch Foundation.

---

[a]Permanent address: University of Hannover, D-3000 Hannover, West Germany.

[b]Permanent address: Free University Berlin, D-1000 Berlin 33, West Germany.

[c]Permanent address: University of Freiburg, D-7800 Freiburg, West Germany, and University of Nevada, Reno, Nev. 89557.

[d]Also at Department of Physics, University of Chicago, Chicago, Ill. 60637.

[e]Permanent address: University of Texas, Austin, Austin, Tex. 78712.

[1]H. W. Hermann and I. V. Hertel, Z. Phys. A **307**, 89 (1982), and references quoted therein.

[2]R. Düren, in *Coherence and Correlation in Atomic Collisions,* edited by H. Kleinpoppen and J. F. Williams (Plenum, New York, 1980), p. 437, and references quoted therein.

[3]T. J. McIlrath, Appl. Phys. Lett. **15**, 41 (1969).

[4]J. L. Le Gouët, J. L. Picqué, F. Wuilleumier, J. M. Bizan, P. Dhez, P. Koch, and D. L. Ederer, Phys. Rev. Lett. **48**, 600 (1982).

[5]F. J. Wuilleumier, in *X-Ray and Atomic Inner-Shell Physics–1982,* edited by B. Crasemann, AIP Conference Proceedings No. 94 (American Institute of Physics, New York, 1982), and references quoted therein.

[6]D. L. Matthews, in *Methods of Experimental Physics,* edited by P. Richard (Academic, New York, 1980), p. 433.

[7]E. Breuckmann, B. Breuckmann, W. Mehlhorn, and W. Schmitz, J. Phys. B **10**, 3135 (1977), and referenees quoted therein.

[8]T. B. Lucatorto and T. J. McIlrath, Phys. Rev. Lett. **37**, 428 (1976).

[9]J. Sugar, T. B. Lucatorto, T. J. McIlrath, and A. W. Weiss, Opt. Lett. **4**, 109 (1979).

[10]V. J. Ross, T. W. Ottley, and V. Pejcev, and D. Rassi, J. Phys. B **9**, 3237 (1976).

[11]H. W. Wolff, K. Radler, B. Sonntag, and R. Haensel, Z. Phys. **257**, 353 (1972).

[12]N. Stolterfoht, in *Proceedings of the International Conference on Inner Shell Ionization Phenomena, Freiburg, 1976,* edited by W. Mehlhorn and R. Brenn (University of Freiburg, Freiburg, Germany, 1976), p. 42–56.