Absolute measurement of photoionization cross sections of excited atoms: Application to determination of atomic beam densities

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A method of simultaneously determining the magnitude of the cross section for photoionization of an excited atom and the atomic density of the ground state is presented. Measurements of σ for $Ba(6s6p\,^1P_1)$, $K(4p)$, and $Na(3p)$ at photon ionizing energy 3.51 eV (3533 Å) are reported and compared with previous values. Application of this technique to the determination of the density in an atomic beam is discussed.

One of the most troublesome problems encountered in atomic beam experiments is the accurate determination of the density of the atomic beam. Several methods have been employed, including the use of surface ionization detectors,[1] measurements of trapped lifetimes,[2] and vapor pressure versus temperature calibrations. We have used all three of these methods in our work, but have continued to search for a more reliable and less time consuming beam density measurement. In this paper we describe such a method. We have applied it to density measurements in beams of Ba, K, and Na. With suitable modification, it could be applied to many other atomic species. In addition to permitting determination of these atomic beam densities, the method also provides absolute values of photoionization cross sections for excited atoms, a subject of considerable recent interest.[3–6]

The method, variations of which have been used[7–9] previously to measure photoionization cross sections but not atom densities, has several attractive features. It is noninvasive in the sense that no elements (e.g., "hot wires"), other than those already present, need be introduced into the apparatus. Also, after initial calibration of the system, *in situ* density determinations can be made in a relatively short time, on the order of 15 min. Another important advantage of this method is that it permits selective measurement of both σ and the atom density for one species in the presence of another, or others, in atomic beam experiments. Since this initial calibration includes an independent measurement of the cross section for photoionization of an excited atomic state, these data should be intrinsically interesting as well as potentially useful in other experiments such as those in which relative photoionization cross sections are determined as a function of photon ionizing energy. These cross sections should also be of interest for comparison with theoretical treatments of photoionization of excited atoms. In this paper we report measurements of the absolute values of three such cross sections.

Our experiments were performed in an apparatus in which an atomic beam was intersected at the center of an electrically shielded cell by two collinear laser beams. One laser beam was used to excite the atoms to the reso-nance level and the other to photoionize the atoms out of that level. The excitation beam, 3 mm diameter and 75 μJ per pulse, was from a grazing incidence dye laser pumped by the frequency doubled 1.06 μm fundamental of a Q-switched Nd-doped yttrium-aluminum-garnet (Nd:YAG) laser, pulse length ~10 nsec. The dye laser beam was passed through a Glan Thompson prism, extinction ratio 10^{-5}, before entering the vacuum system. For each of the three atoms studied, the wavelength of this laser was adjusted to the appropriate resonance line. The ionizing beam was the frequency tripled, 3533 Å, output of the same Nd:YAG laser. Photons of this wavelength have sufficient energy to ionize these excited atoms: $Ba(6s6p\,^1P_1)$, $K(4p)$, and $Na(3p)$.

Ions were swept out of the cell through a small aperture in the side of the cell by an electric field pulse applied ~$\frac{1}{2}$ μsec after the laser pulses. This electric field was perpendicular to the plane of the laser beams and the atomic beam. After emerging from the ion-exit aperture, the ions were detected with a calibrated CuBe particle multiplier and a calibrated charge amplifier.

The uv beam was focused to a diameter much smaller than that of the exciting laser beam by a long focal length lens chosen to make the ionizing volume as nearly cylindrical as possible over the ion collection length as defined by the ion exit aperture. This procedure also minimized problems associated with spatial overlap of the beams. Although both laser beams originate with the same Nd:YAG laser, we nevertheless confirmed the temporal overlap with a photodiode.

Accurate measurement of the atom density using this technique depends upon accurate determination of the interaction volume V defined by overlap of the uv and visible laser beams in the effective collection region. This volume was calculated from the optical properties of the focusing lenses and the geometry of the apparatus. Independent measurements confirmed the calculated dimensions. The effective length of the volume was measured by observing the ion signal as the focal point of the uv laser beam was moved across the ion-exit aperture. It was 0.6 cm. The diameter of the volume was determined for each experiment by measuring the laser spot size using pinhole aper-

tures. This spot size was adjusted for each experiment to avoid saturation of the electronics, but it was always in the range 100–150 μm.

Because the intensity of the excitation laser beam is so high relative to that required to saturate the resonance transition, the Rabi frequency is high enough that spontaneous emission may be ignored during the 10-nsec laser pulse. Furthermore, since the transition is saturated throughout the pulse and the laser light is linearly polarized ($\Delta m = 0$),

$$N_1(t) = N_2(t) = N_T(t)/2 \tag{1}$$

during the laser pulse, where $N_1(t)$ and $N_2(t)$ are the number densities of atoms in the ground and excited states, respectively, at time t and $N_T(t)$ is their sum. Note that Eq. (1) is valid for either fine-structure level of Na and K ($J = \frac{1}{2}$ or $\frac{3}{2}$) because $\Delta m = 0$.

The loss rate by ionization from the resonance state is given by

$$\frac{dN_T(t)}{dt} = -R_i N_2(t) = -R_i \frac{N_T(t)}{2} , \tag{2}$$

where R_i is the rate of photoionization,

$$R_i = I(t)\sigma/\hbar\omega , \tag{3}$$

$I(t)$ is the time-dependent intensity of the 3533-Å (ionizing) laser beam in W/cm^2, σ is the cross section for photoionization of the resonance state to all accessible continuum states at this wavelength, and $\hbar\omega$ is the energy of an ionizing photon 3.5 eV. Integration of Eq. (2) yields

$$N_T(t) = N_0 \exp\left[-\frac{\sigma}{2\hbar\omega}\int_0^t I(t')dt'\right] , \tag{4}$$

where $N_0 = N_T(t = 0)$.

In our experiment, the measured quantity is the total charge Q per set of exciting and ionizing pulses. The total charge per pulse is given by

$$Q/e = \int_0^\tau R_i N_2(t)V dt$$

$$= N_0 V \int_0^\tau \frac{I(t)\sigma}{2\hbar\omega} \exp\left[-\frac{\sigma}{2\hbar\omega}\int_0^t I(t')dt'\right] dt , \tag{5}$$

where e is the electronic charge, V is the interaction volume, and τ is the duration of the laser pulse. Integrating this equation and substituting $Z = (Q/e)$, the total number of ions collected, and

$$\int_0^\tau I(t)dt = U/A , \tag{6}$$

where A is the cross-sectional area of the (very nearly cylindrical) interaction volume V and U is the total energy per uv laser pulse, we have

$$Z = N_0 V\left[1 - \exp\left[-\frac{\sigma U}{2\hbar\omega A}\right]\right] . \tag{7}$$

The pulse energy U was measured using a Scientech Model 38 disk calorimeter.

All quantities in Eq. (7) are either known or measured except N_0 and σ, which were determined by least-squares

TABLE I. Comparison of measured cross sections σ with earlier values.

Atom	σ (Mb)	
	This work	Other work
Na(3p)	3.7 ± 0.7	4.5 ± 1.1 [a]
		4.6 [b]
K(4p)	7.6 ± 1.1	6.7 [b]
Ba($6s6p\ ^1P_1$)	17.6 ± 2.3	12.5 [c]

[a]Reference 10 (experimental and theoretical).
[b]Reference 11 (theoretical).
[c]Reference 12 (theoretical).

fitting the data, Z vs U, to Eq. (7) using a simplex algorithm. Note that $Z \to N_0 V$ as $U \to \infty$, so that for sufficiently high U, as determined by the value at which Z saturates, the determination of N_0 is independent of the photoionization cross section. In other words, N_0 is deduced from the asymptotic value of Z while σ is associated with the shape of the Z vs U data. This results in independent values of N_0 and σ. Note that determination of N_0 is possible only because both transitions are saturated.

Table I is a listing of our measured values of σ for three excited atoms and some previously determined values,[10-12] both experimental and theoretical. Typical data for photoionization of Na(3p) and K(4p) are shown in Figs. 1 and 2 together with the curves calculated by fitting these data to Eq. (7). For sodium, the fitting procedure leads to $N_0 = 10^{11}$ cm^{-3} and $\sigma = 3.7 \pm 0.7$ Mb, the latter in reasonable agreement with our earlier measurement[10] obtained by a different technique. This value of σ is also consistent with theoretical work reported in the same paper. In addition, we find excellent agreement with the calculation of Aymar, Luc-Koenig, and Farnoux.[11] Furthermore, our measurement of σ for K(4p), 7.6 ± 1.1 Mb, is also in excellent agreement with that of Ref. 11. We reiterate, however, that the determination of N_0 does not depend on the derived value of σ so that the density determination does not require prior knowledge of the photoionization cross section. This method of measuring the
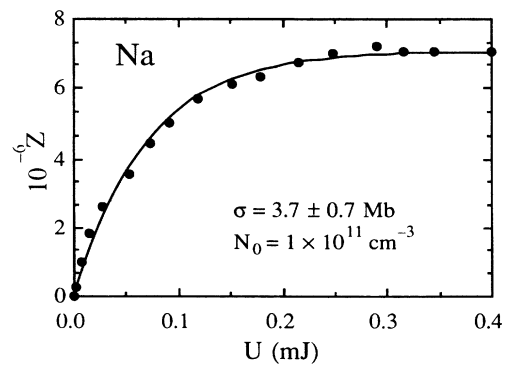

FIG. 1. Photoionization data for Na($3p\ ^2P_{3/2}$). The curve was obtained by fitting the data to Eq. (7) as discussed in the text. The values of the cross section and ground-state density of sodium are obtained from the data.
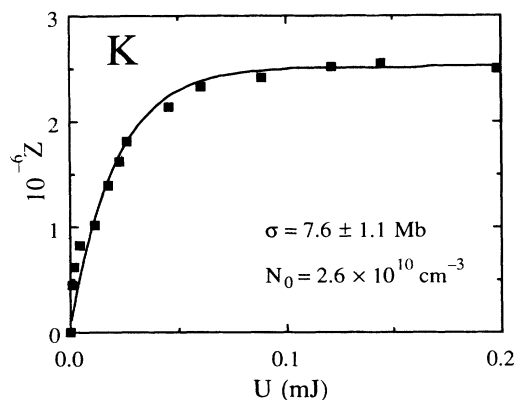
FIG. 2. Photoionization data for $K(4p\,^2P_{3/2})$. The curve was obtained by fitting the data to Eq. (7) as discussed in the text. The values of the cross section and ground-state density of potassium are obtained from the data.
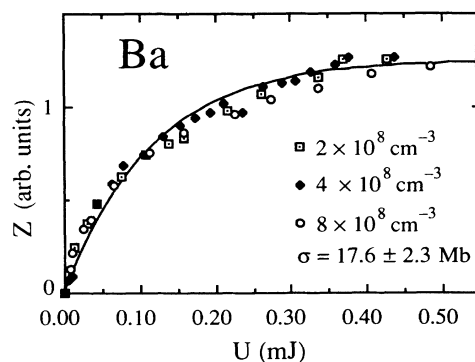

FIG. 3. Data for $Ba(6s\,6p\,^1P_1)$ taken at three different values of the ground-state density as indicated. The value of σ was obtained by fitting all three sets of data to Eq. (7) as discussed in the text. The asymptotic values of each set were normalized.

ground-state density should, therefore, be applicable to a wide variety of experiments.

In addition to the consistency of our measured values of σ with those from previous work, we carefully examined the reproducibility of our data. Figure 3 shows three sets of data for photoionization of $Ba(6s6p\,^1P_1)$ taken at the indicated values of N_0. The cross sections obtained by fitting each set of data are, within error limits, identical to the value obtained by treating all points as one data set.

An interesting aspect of application of this technique to Ba is the possibility of competition between photoionization of the $6p\,^1P_1$ level and amplified spontaneous emission (ASE) to the lower lying (metastable) $5d\,^1D_1$ level. The possibility of ASE was not present in our experiments with Na and K because the laser-excited level was the lowest-lying excited state in each case. The method clearly requires modification if ASE occurs, since depletion of the $6p\,^1P_1$ population would lead to incorrect values of both σ and N_0. ASE will occur when the gain along the length L of the volume defined by the intersection of the laser beams with the Ba beam approaches unity, so that an upper limit to the value of N_0 for which this technique yields reliable results may be obtained by imposing the condition

$$\Delta NL\sigma_{op} \approx 1 ,\qquad (8)$$

where ΔN is the difference in population between the $6p\,^1P_1$ and $5d\,^1D_2$ levels and σ_{op} is the optical cross section. Taking $\Delta N \approx N_0$ and using the known lifetime and branching ratio for the $6p\,^1P_1$ level,[13] this procedure indicates that our method will be reliable for $N_0 \le 10^{11}\,\text{cm}^{-3}$ in Ba.

It is our experience that the biggest source of uncertainty in our measurements lies in the determination of A, the cross-sectional area of the high-intensity uv laser beam. The uncertainties quoted in Table I are thus based on variations in our measurements of A.

The relative orientation of the planes of polarization of the laser beams also affects the measurement of σ. This does not, however, affect the determination of N_0. Since theoretically determined values of σ may be quite final-state specific, comparison with experimental cross sections must be done with the effects of polarization taken into account. To investigate these effects in our experiment we measured σ with the planes of polarization of the excitation laser beam in various directions; a polarizer for the high-intensity uv laser beam was unavailable. The measured cross sections were within 5% of each other for all orientations, which is much smaller than the uncertainty in A. This result indicates a low degree of uv polarization and is consistent with our separate observations of the degree of polarization of the uv laser beam at very low intensity obtained by rotating a polarizer and measuring the laser power. We conclude, therefore, that our measured values of σ, which include s and d continuum final states, are very nearly m-averaged cross sections.

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