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Energy and angular distributions of electrons ejected from the Ba $(6p_j ns_{1/2})_1$ states

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We describe the energy and angular distributions of electrons ejected from the Ba $(6p_j ns_{1/2})_1$ states in their autoionization to the Ba^+ 6*s*, 5*d*, and 6*p* states. The atoms are excited by a three-step laser approach which enables us to excite to the autoionizing state from the spherically symmetric $6sns\ ^1S_0$ states. Since we excite to the discrete part of an autoionizing state, the angular distributions are different from those expected for continuum excitation. Both the angular distributions of the ejected electrons and branching ratios to the available ion states exhibit much more variation than would have been anticipated on the basis of the n^{-3} dependence of the total autoionization rates.

One of the most evident features of autoionizing states is the broadening of spectral lines produced by autoionization, which is easily observed in their optical spectra. The observed spectral broadening is equal to the sum of the autoionization rates to all allowed states of the resulting ion and ejected electron. While the total autoionization rate is itself an important parameter, in cases where more than one final state is available it is interesting for several reasons to know the autoionization rate to each final state. For example, in a calculation of autoionization rates, one must necessarily calculate the rate to each final state of the ion and electron and subsequently sum them to obtain the total autoionization rate. Thus, detailed information regarding the final state of the ion and electron following autoionization allows a more direct comparison with theory. Moreover, in practical applications, such as a laser or a plasma, in which autoionizing states play a role, it is clearly important whether autoionization yields an excited- or ground-state ion.[1]

Here we report what are, to our knowledge, the first energy and angular distribution measurements of the electrons ejected from laser-excited autoionizing Ba atoms. Apart from a detailed analysis of the angular distribution of the electrons from autoionization to final Ba^+ 6*s*, states, we are able to determine the relative amounts of autoionization to the Ba^+ 6*s*, 5*d*, and 6*p* states. These data together with the previously measured total autoionization rates[2] yield the autoionization rates to each final state of the Ba^+ ion. As will be apparent, these measurements show substantial variations in both the angular distributions and branching ratios for autoionization to different ion states which would not have been expected from the smooth n^{-3} variation of the total autoionization rates.[2]

The specific system under study, the Ba $(6p_j ns_{1/2})_1$ states, is illustrated by the $(6p_{1/2}15s_{1/2})_1$ state in Fig. 1. As shown by Fig. 1, for the $(6p_{1/2}15s_{1/2})_1$ state the available odd parity $J=1$ continua are the $6s_{1/2}\epsilon p$ and $5d_j\epsilon p$, ϵf continua, and autoionization of the $6p_{1/2}15s_{1/2}$ state may result in either a $Ba^+ 6s_{1/2}$ ion with a 2.5-eV electron or a $Ba^+ 5d_j$ ion with a ~1.8-eV electron.

As shown in Fig. 1 we use three-step sequential laser excitation of Ba atoms in an atomic beam to autoionizing states coupled with energy and angular analysis of the ejected electrons. After two laser pulses the atoms are in the spherically symmetric $6s15s\ ^1S_0$ state, and only the third laser is involved in the excitation to the autoionizing $(6p_{1/2}15s_{1/2})_1$ state. Thus, we have electric dipole excitation of an unaligned atomic system, a problem identical to photoexcitation from the ground state, and Yang's theorem may be used to predict the angular distribution.[3] Note that this was not the case in the previous multiphoton excitation of Sr through autoionizing states Yang's theorem could not be applied.[4] Because the excitation $6s15s \rightarrow 6p15s$ is essentially the reso-

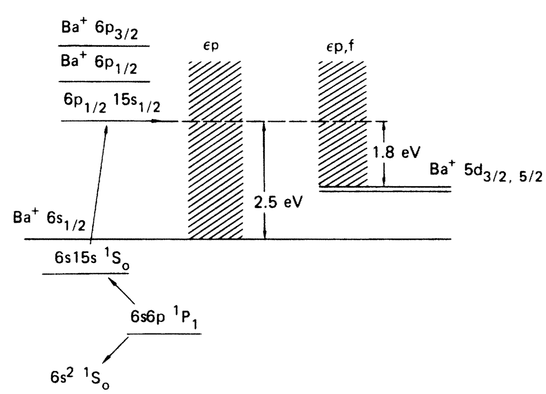

FIG. 1. Energy-level diagram for the excitation of the Ba $(6p_{1/2}15s_{1/2})_{J=1}$ continua above the $Ba^+ 6s$ and 5*d* states are shown as well as the laser pumping steps which are shown as single-ended arrows. Electrons of 2.5 and 1.7 eV are ejected.

nance line of Ba^+ with a spectator $15s$ electron, it is orders of magnitude stronger than the excitation to either of the available continua,[5] and we only excite the autoionizing $(6p_{1/2}15s_{1/2})_1$ level, not the underlying continua.

The atomic beam of Ba is collimated to a diameter of ~ 3 mm and passes midway between two plates 1 cm apart, where it is crossed at 90° by the three colinear laser beams which are focused to $\sim \frac{1}{2}$ mm diameter. The first two lasers are polarized vertically to ensure the production of only the $(6s_{1/2}15s_{1/2})\,{}^1S_0$ state (suppressing the excitation of the nearby ${}^3S_1$ state), and the linear polarization of the third laser is rotated as required by the experiment. Apertures covered by grids are provided in each plate to extract both electrons and ions from the interaction region.

Within a few nanoseconds of the laser pulses, those electrons ejected from the autoionizing atoms in the direction of the slit in the lower plate pass into a 120° cylindrical electrostatic energy analyzer. The analyzer provides an energy resolution of $\Delta E/E = 5\%$ for transmission energies E down to 1 eV (the electrons are accelerated or decelerated to match the transmission energy which is typically 1 to 8 V); the angular resolution defined by the apertures is about 1°. The electrons are detected by a channel electron multiplier.

We measure angular distribution of electrons in the autoionization to a specific state of Ba^+ by observing the electrons ejected at a fixed energy as the polarization of the third laser is rotated. The Ba^+ ions resulting from autoionization are detected by applying a small, ~ 50-V negative voltage pulse to the upper plate 1 μs after the laser pulse to pull the ions into a discrete dynode particle multiplier. Since the number of ions is equal to the total number of atoms excited to the autoionizing state, the ion signal provides a convenient way of normalizing the data to ensure that any variation in the electron signal is not due to an intensity variation coming from the rotation of the third laser polarization, for example.

We have observed the electrons ejected from the $(6p_jns_{1/2})_1$ states for $j = \frac{1}{2}$ and $\frac{3}{2}$ and $n = 9$, 15, 20, and 25. This corresponds to an energy range of about $\frac{1}{2}$ eV in each series. Typical data are shown in Fig. 2 which show the energy spectra of electrons from the $(6p_{1/2}9s_{1/2})_1$ state for $\theta = 0°$ and 90°. Here θ is the angle between the polarization of the third laser and the polar direction in which the electron is ejected.

In all cases, we see angular distributions which can be expressed as

$$I(\theta) = \frac{I_0}{4\pi}[1 + \beta P_2(\cos\theta)] \quad , \tag{1}$$

which is the expected result for electric dipole excitation.[3] Here, I_0 is the total number of electrons ejected in all directions, β is the asymmetry parameter, P_2 is the Legendre polynomial of order 2. To begin, we note that the Cooper-Zare model predicts a $\beta = 2$ (pure $\cos^2\theta$) angular distribution for continuum excitation from a ${}^1S_0$ state to the $6s_{1/2}\epsilon p_j$ continua.[6] However, since we excite the atoms to the $(6pjns_{1/2})_1$ states, not the continuum, we would not expect to find $\beta = 2$ here. In Fig. 3, we show a plot of the

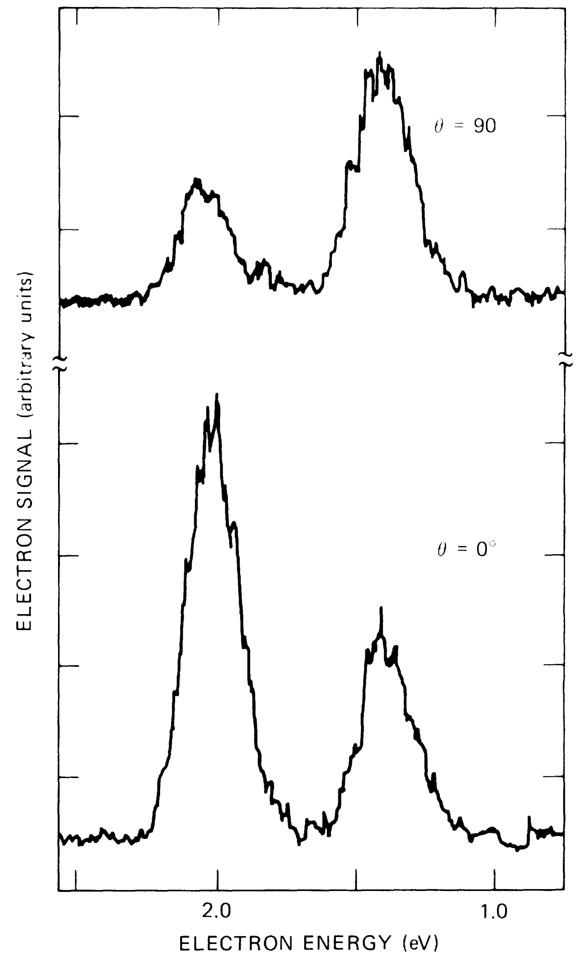

FIG. 2. Typical electron signal from the energy analyzer. The upper and lower traces correspond to $\theta = 90°$ and 0°, respectively, where θ is the angle between the analyzer and laser polarization direction. The two peaks in each trace correspond to the autoionization of $6p_{1/2}9s$ to the $Ba^+ 6s$ and $Ba^+ 5d$ state and result in the ejection of 1.2- or 1.9-eV energy electrons, respectively. While the transmission energy through the electrostatic analyzer is kept fixed at 8 eV, the electrons are accelerated before entering the entrance slit of the energy analyzer. The energy spectrum of the electrons shown here is recorded by slowly increasing the acceleration voltage; the peaks in the spectra appear whenever the sum of the energy of the electron (in eV) and the accelerating voltage (in volts) equals the transmission energy set for the analyzer (8 V for the spectra shown above). Note that the $Ba^+ 5d_{3/2}$ and $Ba^+ 5d_{5/2}$ states are unresolved here.

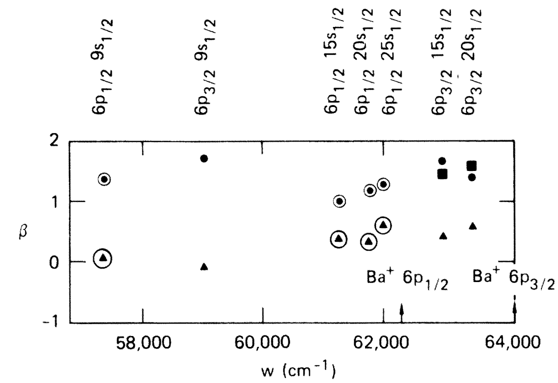

FIG. 3. Asymmetry parameter β for autoionization to the Ba^+6s state (●) and to the pair of Ba $5d$ states (▲) plotted by term energy. The asymmetry parameter for autoionization of $6p_{3/2}ns$ states ($n=15$, 20) to $6p^+_{1/2}$ states is shown by (■). The data points corresponding to $6p_{1/2}ns$ states are circled. The $Ba^+6p_{1/2}$ and $6p_{3/2}$ limits at 62 296 and 63 987 cm^{-1} are indicated as well as the designation of each state.

asymmetry parameters, defined by Eq. (1), measured for electrons from autoionization to the Ba^+6s and $5d$ states. In this work, we only excite the autoionizing state (or resonance), and we see negligible variation in β as we scan the third laser across an autoionizing state. This is, of course, a consequence of the lack of continuum excitation mentioned previously. More importantly, the departure of our observed electron angular distributions from 2 for the autoionization to Ba^+6S state indicates clearly the anisotropy of the electron core interaction at small orbital radius.

We are able to use the observed angular distributions to identify the relative importance of channels through which autoionization occurs, at least for autoionization to the Ba^+6s state. The process may be thought of as follows: The third laser excites the Ba atom to the *jj* coupled $(6p_{1/2}ns_{1/2})_1$ state with $m_j=0$. The *ns* electron, in its highly eccentric orbit, comes, during each orbit, close to the core, where the resulting state is better described in *LS* coupling as $6pns\ ^1P_1$ or $^3P_1$. The *ns* electron is scattered by the core, and there is a finite probability that the electron will be ejected with $l=1$ and the Ba^+ core left in its $6s_{1/2}$ state. We measure the angular distribution of the ejected electron at an orbital radius $r\sim\infty$, where the spin and orbit of the electron are decoupled. Thus, we may describe the final state in terms of l of the electron and the total angular momentum $\vec{J}_{cs}$ of the residual Ba^+ core plus the spin of the outgoing electron.[7] For Ba^+ in the $6s_{1/2}$ state, $J_{cs}=0$ or 1, and these states correspond to the close coupled $^1P_1$ and $^3P_1$ states, respectively. This is a particularly simple case because the resulting Ba^+ state has no orbital angular momentum. Using either the angular momentum transfer approach of Dill and Fano[8] or a more direct approach based on the conservation of total angular momentum, it is a straightforward matter to show that autoionization via the $J_{cs}=0$ and 1 channels leads to $\cos^2\theta$ ($\beta=2$) and $\sin^2\theta$ ($\beta=-1$) angular distributions, respectively. The latter is a good example of a parity-unfavored angular distribution described by Dill and Fano[8] and must therefore occur via a pseudoscalar interaction. The large values of β observed for autoionization to the Ba^+6s state imply that autoionization through $J_{cs}=0$ is roughly three times faster than through $J_{cs}=1$. This is not a surprising result since the spatial overlap of two electron singlet wave functions is better than triplet wave functions.

We note in passing that since the $(6p_{1/2}ns_{1/2})_0$ and $(6p_{3/2}ns_{1/2})_2$ states are pure *triplet* states $^3P_0$, $^3P_2$, the angular distribution measurements reported here in Fig. 3 suggest that their autoionization rates to the Ba^+6s state should be slower, by about a factor of 3, than the $(6p_jns_{1/2})_1$ states. Unfortunately, the large autoionization rates to the Ba^+5d state masks, to some extent, the effect of autoionization to Ba^+6s alone. Further, more detailed investigation of angular distributions of these triplet states are planned.

The branching ratios or fractions of autoionization to each possible state of Ba^+ are easily determined. In Table I we list the observed branching ratios to the possible final ion states. There are two interesting aspects to the branching ratio measurements. First note that a large fraction of the autoionization of the $6p_jns$ states is to excited states of Ba^+. More specifically, it is most interesting that more than half of the autoionization of the $6p_{3/2}ns_{1/2}$ ($n=15,20$) states is to the $6p_{1/2}$ state of Ba^+. This high branching ratio to the $6p_{1/2}$ state of Ba^+ has been utilized recently to generate a Ba^+ laser.[1]

If this proceeds via the $1/r_{12}$ Coulomb repulsion of

TABLE I. Branching ratios for the Ba $(6p_jns_{1/2})_{J=1}$ autoionizing states leading to the $6s^+$ ground and the $5d_j^+$, and $6p^+_{1/2}$ excited state of the ion. The typical statistical uncertainity in the data is ±5%.

Autoionizing state	Branching ratio to on state $6s_{1/2}$ (%)	$5d_{3/2,5/2}$ (%)	$6p_{1/2}$ (%)
$6p_{1/2}9s_{1/2}$	65	35	. . .
$6p_{1/2}15s_{1/2}$	55	45	. . .
$6p_{1/2}20s_{1/2}$	12	88	. . .
$6p_{1/2}25s_{1/2}$	44	56	
$6p_{3/2}9s_{1/2}$	65	35	
$6p_{3/2}15s_{1/2}$	12	15	73
$6p_{3/2}20s_{1/2}$	11	27	62

the two electrons it must be the via quadrupole term in the expansion, which necessarily leads to ϵd electrons. This term apparently dominates dipole contributions to other states of Ba^+, unlike the more familiar case of optical excitations. Such behavior was also suggested by earlier purely spectroscopic measurements.[9]

Second we find a perturbed n dependence in the $6p_{1/2}ns$ series which is indicative of perturbations by the $6p_{3/2}nl$ series leading to large variations in the branching ratios. In particular note that the $6p_{1/2}20s$ state branching ratio to Ba^+6s is anomalously low due, we feel, to interaction with the nearby $6p_{3/2}10d$ state.

These first observations of electron energy and angular distribution from well-defined autoionizing states, excited under single photon absorption conditions, serve to point out several phenomena. We can immediately see the importance of autoionization to excited states of the resulting ion; a process which is of fundamental and applied importance. Second, the observed n dependence of both the angular distribution and the branching ratio yields valuable information for calculations on the dynamics of the autoionization process. Finally we are, in favorable cases, able to determine the close coupled channels through which autoionization occurs.

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