Experimental determination of the total angular momentum of autoionizing states of the 6p7p configuration of the Ba atom by polarization laser spectroscopy

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Two-step excitation of autoionizing states of the $6p7p$ configuration of the Ba atom has been used to identify these states in terms of the total angular momentum J. The identification procedure makes use of selection rules for stepped excitation by polarized radiation. Spectra of the linear and circular dichroism along the profile of the autoionizing resonance are reported. Theoretical expressions are derived for these properties. These expressions are used along with the experimental data in the resonance region to calculate two theoretical parameters, the ratios of the squares of the dipole matrix elements for transitions from the excited state.

1. INTRODUCTION

Autoionizing resonances of atoms have been studied actively, both experimentally and theoretically, in recent years. Substantial progress has been achieved in this field thanks to improvements in laser-spectroscopy methods and also in the quality of the theoretical models. The agreement between experiment and theory is best for autoionizing resonances which lie well above the boundary of the continuous spectrum, and for which the quantum-defect method can be used successfully.[1] Calculations for autoionizing resonances near the ionization limit of the atom are carried out in the Hartree–Fock–Slater approximation.[2] In this case the agreement with experimental data is often not satisfactory. It thus becomes necessary to develop experimental methods for studying autoionizing resonances. The possibility of extracting such theoretical parameters as a ratio of dipole matrix elements from experimental data becomes very important for the derivation of a theory. In the present paper we identify autoionizing resonances of the $6p7p$ configuration of the Ba atom. We also report measurements of the linear and circular dichroism of an autoionizing resonance with angular momentum $J=1$ (the excitation energy is 52158 cm^{-1}). It thus becomes possible to determine two theoretical parameters, namely the ratios of the dipole matrix elements for transitions from the $6s8p$ ($^1P_1$) excited state to the d and s continua.

2. EXPERIMENTAL SETUP

The experimental setup has been described in detail previously.[3–5] We will restrict the discussion here to a summary of the basic parameters. The Ba atoms were excited and ionized by laser light which entered a vacuum chamber evacuated to a pressure of $5\cdot 10^{-7}$ torr (Fig. 1). A beam of Ba atoms produced by an effusion source was formed in the region of the interaction with the laser light. The geometry of the beam was set by a diaphragm of diameter $d_1=2$ mm at the exit from the source, 15 mm from the interaction region. The diameter of the beam of Ba atoms in the interaction region was estimated to be 10 mm. The density of atoms in the beam in the interaction region was 10^9 atoms/cm^3.

Two pulsed laser beams entered the interaction chamber to excite and ionize the Ba atoms. These beams made an angle of 180° with each other and were oriented perpendicular to the direction of the flux of Ba atoms from the source. The second harmonic of an LTI-405 YAG laser was used to pump two dye lasers. The home-brew dye lasers had an output line with a spectral width of 2 cm^{-1}, a frequency scanning interval of 550–680 nm, and a energy of 5 mJ per pulse. The wavelength of the light from one of the dye lasers was doubled in a KDP crystal. The linear polarization of the light was selected with the help of a Glan prism. The polarization was rotated with the help of a double Fresnel rhomb. The degree of linear polarization was 95%, and the degree of circular polarization 80%.

Polarization spectra of the autoionizing resonances were measured by a time-of-flight mass spectrometer. The ion source of this spectrometer had ion-accelerating fields $U_1=1.5$ kV/cm and $U_2=200$ V/cm. The spectrometer was synchronized by the laser pulse. The aperture of the ion packet was about 12 mm. The mean free path of this packet was 80 cm. As detectors we used two microchannel plates 46 mm in diameter, together forming a chevron configuration. The amplitudes of the ion and light signals were measured by a V9-5 pulsed digital voltmeter. An average was taken over 50 laser shots to find each point in the spectrum.

3. THEORY

Let us take a closer look at the two-step ionization of atoms. The laser used in the first step, which excites a discrete transition, creates a ensemble of polarized atoms in an excited state. This is an aligned state[7] if the light from the first laser is linearly polarized, while it is an oriented state if the light is circularly polarized. The laser used in the second step ionizes the polarized excited state. The photoionization cross section of the polarized atom depends on the direction and type of polarization of the light from the second laser. The difference between the photoionization cross sections for right- and left-hand-polarized light (of the second laser) is the "circular dichroism." The

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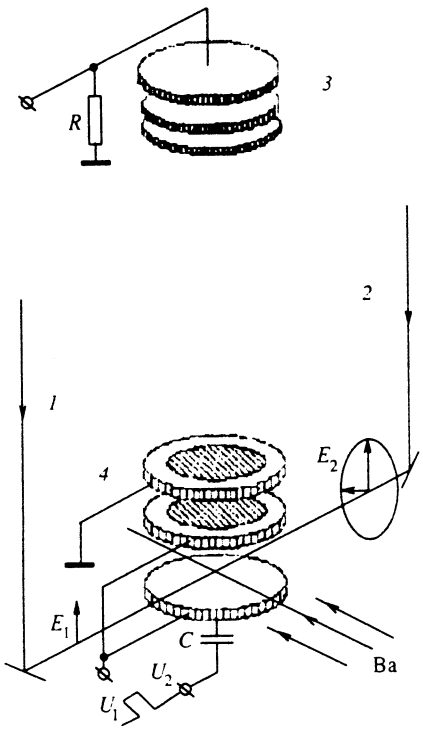

FIG. 1. Experimental layout. *1, 2*—Light from two dye lasers; *3, 4*—detector and ion source of the time-of-flight mass spectrometer.

difference between the ionization cross sections for linearly polarized light with mutually perpendicular polarizations is the "linear dichroism."

The photoionization of a polarized atom by light of arbitrary polarization was studied in Refs. 8 and 9. Using the formalism developed in those studies, we derive an expression for the photoionization cross section of a polarized atom in the case of interest here, in which the first laser excites the discrete $6s8p$ state ($^1P_1$; 35,892 cm^{-1}). The second laser ionizes this state. The *jj* approximation coupling is valid for excited states.[10] Analysis shows that the energy levels of the $6s8p$ configuration[11,12] correspond specifically to *jj* coupling. The wave function of the $6s8p$ state can thus be written in general as

$$\Phi_i=\Phi_{LSJM}=\sum_{m_j,M_j'}(-1)^{J'-j-M}(2J+1)^{1/2}\begin{pmatrix} j & J' & J \\ m_j & M_J' & -M\end{pmatrix}\phi_{nljm_j}\Phi_{L'S'J'M'}. \quad (1)$$

The wave function of the final state, in which there is an ion and a photoelectron, can be expanded in states corresponding to a definite total angular momentum of the system consisting of the ion and the photoelectron:

$$\Phi_f=\phi_{\varepsilon l_1 j_1 m_j^1},\quad \Phi_{L'S'J'M_J'}$$

$$=\sum_{J_1M_J^1}(j_1J'm_j^1M_J'|J_1M_J^1)\Phi_{J_1M_J^1}$$

$$=\sum_{J_1,M_J^1}\sum_{m_j',M_J''}(2J_1+1)\begin{pmatrix} j_1 & J' & J_1 \\ m_j^1 & M_J' & -M_J^1\end{pmatrix}\times\begin{pmatrix} j_1 & J' & J_1 \\ m_{j1}' & M_J'' & -M_J^1\end{pmatrix}\times\phi_{\varepsilon l_1 j_1 m_j'}\Phi_{L'S'J'M_J''}, \quad (2)$$

where ε is the energy of the photoelectron. Using these wave functions, we find the dipole matrix element:

$$\langle j_1 m_{j1}, J'M_J'|d_\lambda|JM\rangle$$

$$=\sum_{J_1M_J'}(-1)^{J'-j_1-M_J'}[J_1]^{1/2}\begin{pmatrix} J_1 & J' & J_1 \\ m_{J1} & M_J' & -M_J^1\end{pmatrix}\times\langle J_1M_J^1|d_\lambda|JM\rangle, \quad (3)$$

where $[J_1]\equiv 2J_1+1$. Making use of the Wigner–Eckart theorem[10]

$$\langle J_1M_J^1|d_\lambda|JM\rangle=\begin{pmatrix} J_1 & 1 & J \\ -M_J^1 & \lambda & M\end{pmatrix}\langle J_1\|\, d\|\, J\rangle, \quad (4)$$

We obtain

$$\langle J_1\|\, d\|\, J\rangle=[J,J_1,j,j_1]^{1/2}(-1)^{J+J'-j-j_1+1/2}\begin{Bmatrix} j_1 & 1 & j \\ l & \frac{1}{2} & l_1\end{Bmatrix}\times\begin{Bmatrix} j_1 & 1 & j \\ J & J' & J_1\end{Bmatrix}\langle \varepsilon l_1 j_1\|\, d\|\, nlj\rangle, \quad (5)$$

where $\varepsilon l_1 j_1\|\, d\|\, nlj$ is the usual reduced dipole matrix element[13]:

$$\langle \varepsilon l_1 j_1\|\, d\|\, nlj\rangle=[l,l_1]^{1/2}(-1)^{l_1}\begin{pmatrix} l_1 & 1 & l \\ 0 & 0 & 0\end{pmatrix}\int_0^\infty R_{\varepsilon l_1}^{j_1}(r)R_{nlj}(r)r^3dr. \quad (5a)$$

It is convenient to write the general expression for the photoionization cross section of a polarized atom for light of arbitrary polarization in terms of the density matrices as follows:

$$\sigma(\mathbf{n})=4\pi^2\alpha\omega\sum_{\lambda,\lambda'}\sum_{M,M'}\sum_{m_{j1},M_J'}\langle j_1m_{j1},J'M_J'|d_\lambda|JM\rangle\times\langle JM|\rho_a|JM'\rangle\langle\lambda|\rho^\gamma|\lambda'\rangle\times\langle JM'|d_{\lambda'},|j_1m_{j1},J'M_J'\rangle. \quad (6)$$

Here α is the fine-structure constant, and ω is the energy of the photons (we are using the atomic system of units, with $\hbar=m=e=1$). In the laboratory frame of reference, whose z axis coincides with the direction of the photon beam, the density matrix of the photons is[7]

$$\langle\lambda|\rho^{\gamma}|\lambda'\rangle=1/2\begin{pmatrix}1+\xi_2 & -\xi_3+i\xi_1\\ -\xi_3-i\xi_1 & 1-\xi_2\end{pmatrix}$$

$$=\sum_{k,\kappa}(-1)^{1-\lambda}[k]^{1/2}\begin{pmatrix}1 & 1 & k\\ \lambda & -\lambda' & -\kappa\end{pmatrix}\rho^{\gamma}_{k\kappa}, \quad (7)$$

where ρ^{γ} are the multipoles of the states, and ξ are the Stokes parameters. The Stokes parameter $\xi_2=+1$ (-1) corresponds to right-hand (left-hand) circular polarization of the photon, $\xi_3=+1$ (-1) corresponds to linear polarization along the $x(y)$ axis, and $\xi_1=+1$ (-1) corresponds to linear polarization at an angle of 45° (135°) with the x axis.

We assume that the atoms in the excited state are polarized in some direction $\mathbf{n}$ ($\mathbf{n}$ is a unit vector) other than the z axis of the laboratory frame of reference. In other words, the density matrix of the atoms, $\rho^{\mathbf{n}}_a$, is diagonal in the primed coordinate system whose z' axis runs along $\mathbf{n}$. The density matrix in the laboratory frame is then conveniently expressed in terms of the diagonal matrix in the primed frame by means of the finite-rotation matrix[7,14] [Wigner D-functions $D^{j}_{mm'}(\omega)$, where ω is the set of Euler angles describing the rotation from the unprimed frame of reference to the primed one]:

$$\langle JM|\rho_a|JM'\rangle=\sum_{M_1}D^{J}_{MM_1}(\omega)$$

$$\times\langle JM_1|\rho^{\mathbf{n}}_a|JM_1\rangle D^{J*}_{M_1M'}(\omega). \quad (8)$$

Again introducing the multipoles of the states, $\rho^{\mathbf{n}}_{N0}$, we find[7]

$$\langle JM|\rho_a|JM'\rangle=\sum_{N,M_N}\sqrt{4\pi}(-1)^{J-M}$$

$$\times\begin{pmatrix}J & J & N\\ M & -M' & -M_n\end{pmatrix}Y^{*}_{NM_N}(\mathbf{n})\rho^{\mathbf{n}}_{N0}, \quad (9)$$

where

$$\rho^{\mathbf{n}}_{N0}=\sum_{M_1}[N]^{1/2}(-1)^{J-M_1}\begin{pmatrix}J & J & N\\ M_1 & -M_1 & 0\end{pmatrix}$$

$$\times\langle JM_1|\rho^{\mathbf{n}}_a|JM_1\rangle. \quad (10)$$

Substituting expressions (3), (7), and (9) into (6), and summing over the projections of the angular momenta, we finally find a general expression for the photoionization cross section of polarized atoms:

$$\sigma(\mathbf{n})=4\pi^2\alpha\omega\sum_{J_1}\sum_{k\kappa}\sqrt{4\pi}[k]^{-1/2}Y^{*}_{k\kappa}(\mathbf{n})\rho^{\mathbf{n}}_{k0}\rho^{\gamma}_{k\kappa}$$

$$\times(-1)^{J+J_1+1}\begin{Bmatrix}1 & 1 & k\\ J & J & J_1\end{Bmatrix}|\langle J'l_1j_1J_1\|d\|Jlj\rangle|^2. \quad (11)$$

We turn now to a particular process, which is studied experimentally. We consider the case in which the output from the first laser is linearly polarized in the x direction, and its photon energy is equal to the transition energy of the barium atom to the $6s8p(^1P_1)$ excited state. We then have $J=1$ and $M_1=0$; for the multipoles of the atomic states, (10), we find

$$\rho^{n}_{00}=1/\sqrt{3}, \quad \rho^{n}_{10}=0, \quad \rho^{n}_{20}=-\sqrt{2/3}. \quad (12)$$

Let us assume that the output of the second laser is also linearly polarized in the x direction. Corresponding to this case are the photon state multipoles[7]

$$\rho^{\gamma}_{00}=1/\sqrt{3}, \quad \rho^{\gamma}_{20}=1/\sqrt{6}, \quad \rho^{\gamma}_{2\pm2}=-1/2. \quad (13)$$

Substituting them into (11), we find

$$\sigma_{\parallel}=\frac{4\pi^2\alpha\omega}{9}\left(3d_0^2+\frac{6}{5}D_2^2\right), \quad (14)$$

where for simplicity we use the notation $Dj_1=\langle J_1\|d\|J\rangle$. Similarly, when the output of the second laser is linearly polarized in the y direction ($\rho^{\delta}_{2\pm2}=1/2$), we find

$$\sigma_{\perp}=\frac{4\pi^2\alpha\omega}{9}\left(\frac{3}{2}D_1^2+\frac{9}{10}D_2^2\right). \quad (15)$$

From an experimental standpoint, it is convenient to replace the linear dichroism found above by the normalized linear dichroism defined by[15,16]

$$A_{\mathrm{LD}}=\frac{\sigma_{\perp}-\sigma_{\parallel}}{\sigma_{\perp}+\sigma_{\parallel}}=\frac{-2D_0^2+D_1^2-0.2D_2^2}{2D_0^2+D_1^2+1.4D_2^2}. \quad (16)$$

Correspondingly, we consider the case of circularly polarized light. We assume that the light from the first-step laser is circularly polarized in the z direction, so it generates an excited state of the atom with $J=1$, $M_1=+1$. We then have

$$\rho^{n}_{00}=1/\sqrt{3}, \quad \rho^{n}_{10}=1/\sqrt{2}, \quad \rho^{n}_{20}=1/\sqrt{6}. \quad (17)$$

The light from the second-step laser is also circularly polarized in the z direction, so we have

$$\rho^{\gamma}_{00}=1/\sqrt{3}, \quad \rho^{\gamma}_{20}=1/\sqrt{6}, \quad \rho^{\gamma}_{10}=\pm1/\sqrt{2}, \quad (18)$$

where the $\pm$ in the equation for ρ^{γ}_{10} correspond to right- and left-hand circular polarizations. For these values of the multipoles of the states, the general expression (11) yields

$$\sigma_{++}=\frac{4\pi^2\alpha\omega}{9}D_2^2 \quad (19)$$

for two identical circular polarizations and

$$\sigma_{+-}=\frac{4\pi^2\alpha\omega}{9}\left(2D_0^2+\frac{1}{2}D_1^2+0.1D_2^2\right) \quad (20)$$

for two opposite circular polarizations. We thus find an expression for the normalized circular dichroism:

$$A_{\mathrm{CD}}=\frac{\sigma_{++}-\sigma_{+-}}{\sigma_{++}+\sigma_{+-}}=\frac{-2D_0^2-D_1^2+D_2^2}{2D_0^2+D_1^2+1.4D_2^2}. \quad (21)$$

Expressions (16) and (21) thus express quantities that can be measured experimentally in terms of reduced dipole matrix elements that can be calculated theoretically.

TABLE I.

E, cm^{-1}	Identification suggested in Ref. 18	Present study; excitation through the intermediate resonance, (J)		
		$6s6p(^1P_1)$	$6s7p(^1P_1)$	$6s8p(^1P_1)$
50383	$^1P_1$ $(1/2,1/2)_1$		2	
51113	$^3D_1$ $(1/2,3/2)_1$	2		2
51494	$^3P_0$ $(1/2,1/2)_0$	2		2
52158	$^3P_1$ $(3/2,1/2)_1$	1		1
52583	$^3P_1$ $(3/2,3/2)_1$			1

4. EXPERIMENTAL PROCEDURE AND RESULTS

An autoionizing resonance of the $6p7p$ configuration in Ba atoms was first studied in Ref. 18. Eight resonances were found experimentally via two-step excitation from the $6s5d$ state. These resonances were identified on the basis of calculations carried out in the Slater–Condon approximation. The results of those calculations are shown along with experimental results in Table I. The positions of the same autoionizing resonances were found in Ref. 2 by the Hartree–Fock–Dirac method, with superposition of configurations being taken into account. The results of the calculations carried out in Refs. 2 and 18 are extremely different. They also differ from our own identification carried out in a previous study,[4,5] by a method based on selection rules for the two-step excitation by polarized light through the intermediate states $6s7p(^1P_1)$ and $6s8p(^1P_1)$.

In the present study we use excitation of the same autoionizing resonances through the intermediate resonances $6s6p(^1P_1)$, $6s7p(^1P_1)$, and $6s8p(^1P_1)$.

Let us examine each of these cases in more detail.

a. Excitation of autoionizing resonances through the $6s7p(^1P_1)$ and $6s8p(^1P_1)$ intermediate resonances. Figures 2 and 3 show measurements of the photoionization cross sections of barium atoms for various combinations of the polarizations of the laser light, for the resonance with $E=52{,}158$ cm^{-1}, $J=1$:

$$6s^2(^1S_0) \xrightarrow[(1)]{} 6s8p(^1P_1) \xrightarrow[(2)]{} 6s6p(J=0;1;2).$$

The $6p7p$ (52,158 cm^{-1}) resonance that we studied has total angular momentum[5,19] $J=1$. It follows from the selection rules[5] that this resonance should not occur if the beams from the two lasers, (1) and (2), are linearly polarized in the same plane or if they are circularly polarized in opposite directions (recall that they are counterpropagating). It can be seen from Fig. 2 that this condition holds well in the case of parallel linear polarizations, while a resonance is still seen in the case of the corresponding circular polarizations (Fig. 3). This result suggests that the degree of circular polarization of the light is not 100%. On the basis of the results in Figs. 2 and 3, we calculated the normalized linear dichroism and normalized circular dichroism from expressions (16) and (21). Figure 4 shows the results of these calculations for the case of the linear dichroism. The width of the resonance in the dichroism curves is considerably greater than the width in the photo-

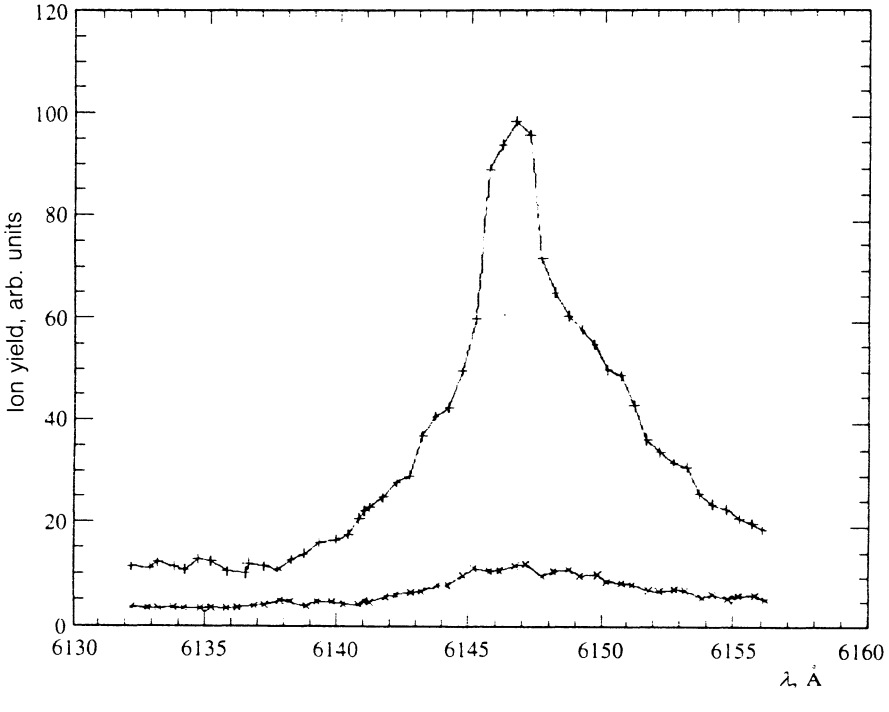

FIG. 2. Shape of the $6p7p$ autoionizing resonance ($E=52{,}158$ cm^{-1}, $J=1$) versus the wavelength of the output from the laser used in the second step [for the $6s8p$ $(^1P_1)$ intermediate resonance]. The beams from the two lasers are linearly polarized, either in mutually perpendicular planes (+) or in parallel planes (×).

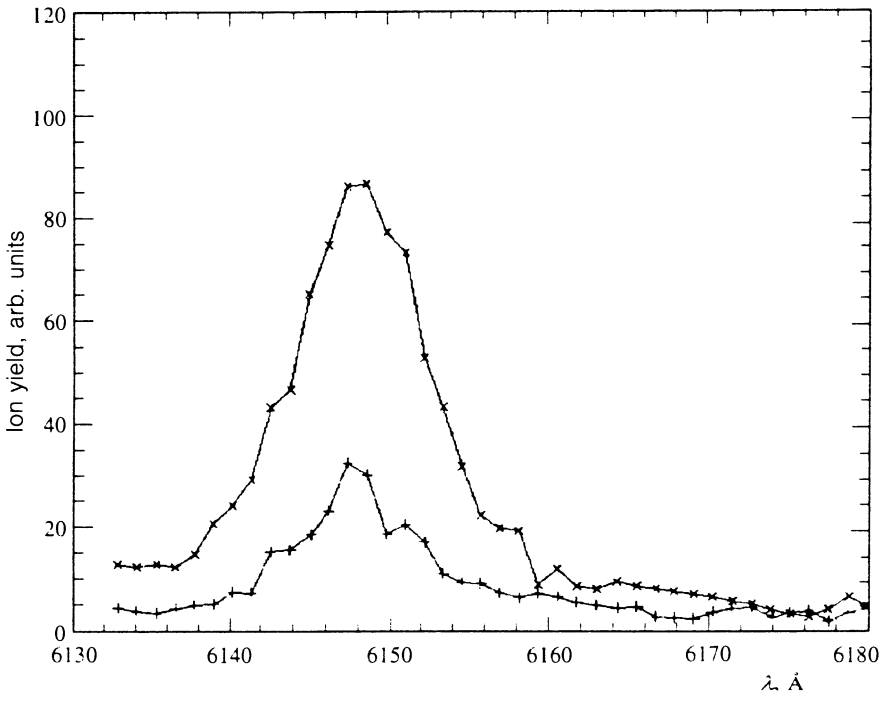

FIG. 3. Shape of the $6p7p$ autoionizing resonance ($E=52{,}158$ cm^{-1}, $J=1$) versus the wavelength of the output from the second-step laser [for the $6s8p$ ($^1P_1$) intermediate resonance]. The beams from the two lasers are circularly polarized, either in the same direction ($\times$) or in opposite directions ($+$).

ionization cross section, as in the case of polarization parameters.[19] With two independent measurable quantities available, A_{CD} and A_{LD}, we can determine only two theoretical parameters, while expressions (16) and (21) contain five different dipole matrix elements. However, it is not difficult to see that these matrix elements appear in certain combinations, so that there are actually only three independent quantities: d_{10}^2 and two linear combinations which we write as

$$\nu_1^2 = D_1^2/D_0^2, \tag{22}$$

$$\nu_2^2 = D_2^2/D_0^2 \tag{23}$$

we find

$$A_{CD} = \frac{-2-\nu_1^2+\nu_2^2}{2+\nu_1^2+1.4\nu_2^2}, \tag{24}$$

$$A_{LD} = \frac{-2+\nu_1^2-0.2\nu_2^2}{2+\nu_1^2+1.4\nu_2^2}. \tag{25}$$

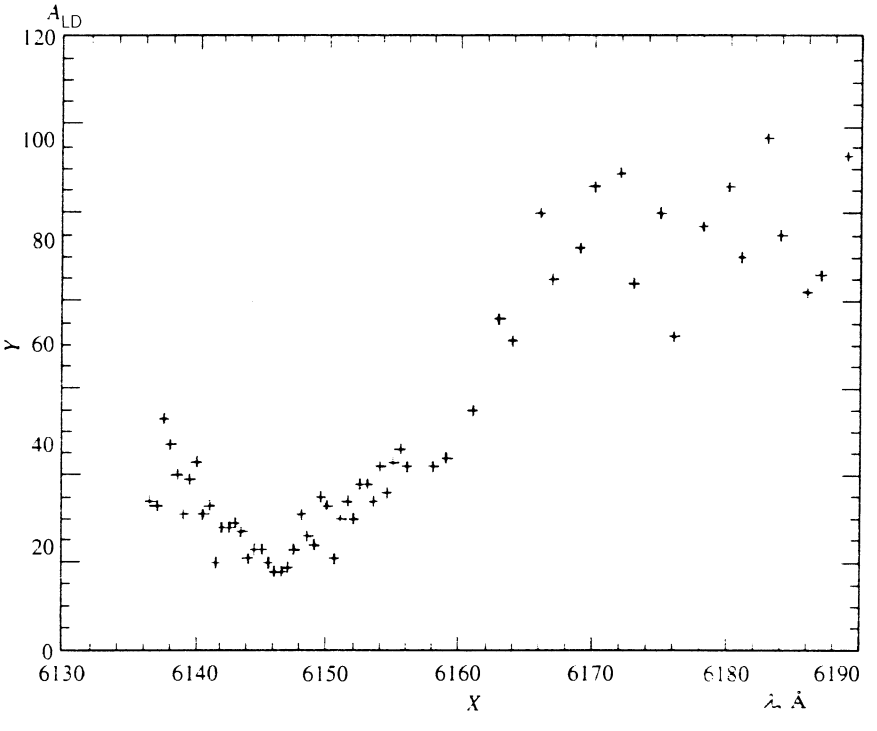

FIG. 4. Linear dichroism of the autoionizing resonance $6p7p$ ($E=52{,}158$ cm^{-1}) versus the wavelength of the second-step laser.

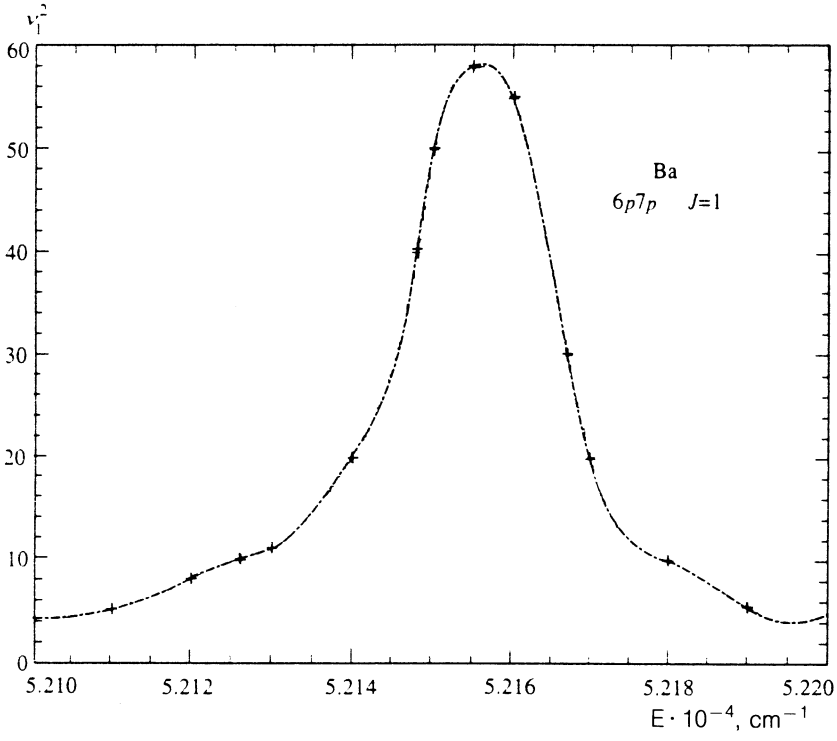

FIG. 5. Behavior of the parameter ν_1^2 [see (23)] along the line of the autoionizing resonance of the $6p7p$ ($J=1$) configuration as found from the measured values of A_{LD} and A_{CD}.

From the measured quantities A_{CD} and A_{LD} we can thus extract the two parameters ν_1^2 and ν_2^2 for each photon energy. Expressions (24) and (25) were derived for 100% light polarized. For a degree of polarization other than 100%, it is a straightforward matter to derive corresponding expressions for A_{CD} and A_{LD}, by making use of the general expression for the photoionization cross section in terms of the multipoles of the states (11). However, the corresponding equations are more complicated, so we will not write them out here. In the very simple case in which the two laser beams can be characterized by the same values of the Stokes parameters ($\xi_1=\xi_3=0$, $0<\xi_2<1$), the circular dichroism turns out to be proportional to ξ_2^2. We made use of this proportionality in analyzing the experimental results. Using the data on A_{LD} and A_{CD} in Fig. 4, we found values of the dimensionless parameters ν_1^2 and ν_2^2. The first of these parameters is shown in Fig. 5. The second varies slightly near the observed resonance with total angular momentum $J=1$, since it is the ratio of dipole matrix elements corresponding to transitions to states with total

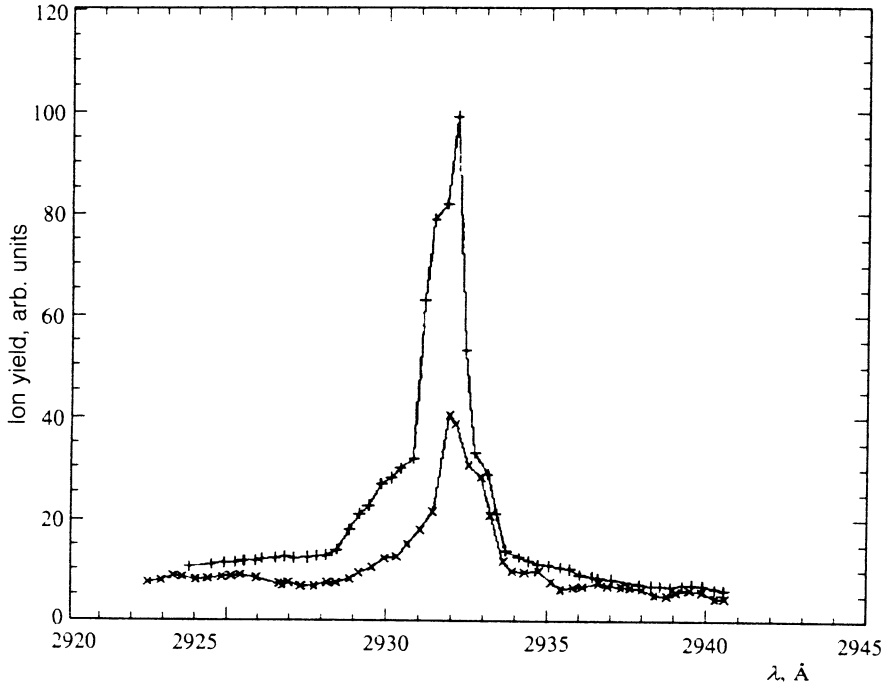

FIG. 6. Shape of the $6p7p$ autoionizing resonance ($E=52{,}158$ cm^{-1}, $J=1$) versus the wavelength of the second-step laser [with the $6s6p$ ($^1P_1$) intermediate resonance]. The beams from the two lasers are linearly polarized, either in mutually perpendicular planes (+) or in parallel planes (×).

angular momenta $J=2$ and $J=0$. We found the value $\nu_2^2=1.3\pm0.3$. Unfortunately, no theoretical calculations have been carried out for this process so far.

b. Excitation of autoionizing resonances through the $6s6p(^1P_1)$ intermediate state

$$6s^2(^1S_0) \xrightarrow[(1)]{} 6s6p(^1P_1) \xrightarrow[(2)]{} 6s6p(J=0;1;2).$$

The power density of the first-step laser (1) is 10^7 MW/cm^2, which saturates the given transition. No two-photon ionization from the $6s6p$ state due to light from the first-step laser was observed. This result is attributed to the low power density of the light and also to the circumstance that the Rabi frequency (Ω) for the induced $6s6p\rightarrow 6s^2$ transition increases more rapidly than the probability for two-photon ionization from the $6s6p(^1P_1)$ state as the laser power density is raised. As a result, there is a decrease in the cross section for two-photon ionization from the $6s6p$ state.

It was possible to eliminate the effect of two-photon ionization from the $6s^2(^1S_1)$ ground state by the light from laser (2) by reducing the power density of this light to 10^5 W/cm^2.

Figure 6 shows measurements of the photoionization cross section for autoionizing resonances in the $6p7p$ configuration of the Ba atom with total angular momentum $J=1$. Table I shows measurements of the total angular momentum of the autoionizing resonances for other resonances.

5. CONCLUSION

The error in the determination of the parameters ν_1^2 and ν_2^2 is still large, as can be seen from the scatter in the experimental points in Figs. 2–5. The reason is that there are instabilities in the intensity and frequency of the laser used to pump the intermediate state; the ion signal is unstable as a result.

In summary, the use of polarized laser light makes it possible to extract two theoretical parameters from experimental data without measuring angular distributions of photoelectrons. This is an important step toward a completely quantum-mechanical experiment,[19] in which it is necessary to determine five independent parameters in order to completely describe the photoionization process. The next step, which will make it possible to extract more information from an experiment, is to measure the angular distributions of the photoelectrons. Such measurements constitute a far more difficult problem.

We are deeply indebted to N. B. Delone for sustained interest in this study.

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