Angular distribution of photoelectron emission as a result of three-photon ionization of Ba atoms

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The angular distribution of photoelectrons produced by ionization of Ba atoms was investigated. The atoms were excited into the $6p^2(^1S_0)$ state of the Ba atom by a two-photon process with linearly polarized dye-laser radiation. This state was ionized by radiation from the same laser. The rotation of the polarization plane of the radiation made it possible to investigate the angular distribution of the photoelectrons for a fixed position of the time-of-flight electron-energy analyzer. The parameter describing the anisotropy of the angular distribution of the emission of photoelectrons was determined by approximating the experimentally obtained angular dependence, which corresponds to the ground state of the ion, of the electronic signal.

1. During the last ten years the anisotropy of the angular distribution of photoelectrons has been used to study the ionization of atoms.[1-3] Of special interest in this study is the possibility of performing a complete quantum-mechanical photoionization experiment. To perform such an experiment, it is necessary to measure the angular distribution and the spin orientation of the photoelectrons. At present, the measurement of the spin of photoelectrons is a complicated experimental problem involving a large experimental error.

In Ref. 4 a method was proposed for performing a complete quantum-mechanical photoionization experiment based on measurements of the angular and energy distributions of electrons for optically oriented atoms. Such experiments can be performed with a two-step process of excitation with polarized radiation and they are now used to study photoionization.[5-7]

In the present paper we report the results of an experimental study of the angular distribution of electrons with ionization of Ba atoms from the excited state $6p^2(^1S_0)$. The excitation scheme of the Ba atoms is shown in Fig. 1.

2. Three-photon ionization of Ba atoms was investigated in a vacuum chamber, evacuated to a pressure of 5×10^{-7} torr. A beam of Ba atoms from an effusion source was formed in the region of interaction with the laser radiation. The geometry of the atoms of the beam was determined by a diaphragm with diameter $d_1 = 2$ mm. The diaphragm placed at the exit of the source 40 mm from the region of interaction with the laser radiation. The diameter of the laser beam in the region of interaction with the Ba atoms

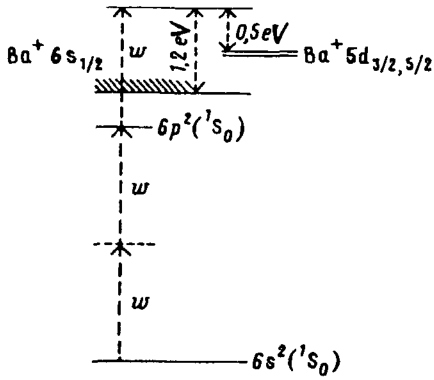

FIG. 1. Energy levels of the Ba atom and the transitions employed.

was estimated to be 0.5 mm. The concentration of atoms in the beam in the region of interaction with the laser radiation was equal to 10^{11} atoms/cm^3.

The Ba atoms were excited and ionized by pulsed dye-laser radiation, oriented perpendicular to the direction of the beam of Ba atoms. The second harmonic of the IZ-25 YAG laser was used as a master oscillator to pump the dye laser. The parameters of our hand-made dye laser[8] are as follows: The spectral width of the lasing line was 2 cm^{-1}, the frequency scanning range was 550–680 nm, the radiation pulse energy was 1.5 mJ, and the laser pulse duration was 20 ns. The linear polarization of the radiation was obtained with a Glan prism. A double Fresnel's rhomb was used to rotate the polarization. The degree of linear polarization was 98%.

The region of interaction of the atoms with the laser radiation was screened from the earth's magnetic field with a triple permalloy screen, each screen being 2 mm thick. The residual magnetic field in the region of electron drift was 2 mHz. The free-drift length of the electrons was 10 cm. The diameter of the diaphragm giving an angular resolution of $\pm 5°$ was equal to 2 mm. A S7-19 oscillograph with a 5-GHz band served as a recording system. The signal was recorded on photographic film. The construction of the time-of-flight electron-energy analyzer is described in Ref. 9.

3. Three-photon ionization of the Ba atoms was achieved by the following scheme:

$$\mathrm{Ba}[6s^2(^1S_0)] \xrightarrow{2\omega(581.9\ \mathrm{nm})} \mathrm{Ba}^*[6p^2(^1S_0)], \qquad (1)$$

$$\mathrm{Ba}^*[6p^2(^1S_0)] \xrightarrow{\omega(581.9\ \mathrm{nm})} \mathrm{Ba}^+[6s(^2S_{1/2})],\ 5d(^2D_{3/2,5/2})+e^-[\epsilon p_{1/2},\epsilon f_{3/2},\epsilon f_{5/2}].$$

The energy corresponding to three-photon ionization in the continuous spectrum coincides with the position of the edge of the contour of the autoionization resonance with the configuration $6p8s(^3P_1)$.[10] This greatly complicates the theoretical analysis of the ionization process.

The use of linearly polarized radiation at the first step of excitation made it possible to produce an ensemble of oriented atoms in the state $6p^2(^1S_0)$. The oriented atoms in the excited state can be depolarized by the dipole–dipole interaction and effusion of

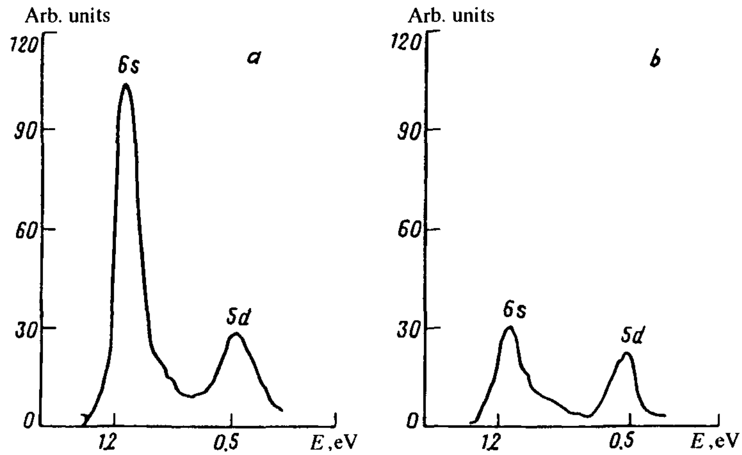

FIG. 2. Electronic signal obtained from the energy analyzer for two values of the angle θ between the axis of the electron-energy analyzer and the direction of polarization of the laser radiation: a — 0° and b — 90°.

radiation. The advantage of two-photon excitation over a two-step process,[11] which can be realized via the intermediate state $6s6p(^1P_1)$, is that there is no depolarization of the ensemble of oriented atoms, since a one-photon transition into the ground state is forbidden.

The angular distribution of the photoelectrons which correspond to the $6s$ state of the ion was measured by rotating the polarization plane of the radiation. Because of the isotropy of the orientation of the $6p^2(^1S_0)$ state, the analytical expression for the angular distribution of the electrons has the following form:[12]

$$\frac{d\sigma}{d\theta}=\frac{\sigma_s}{4\pi}[1+\beta\, P_2(\cos\theta)], \tag{2}$$

where σ_s is the total cross section, β is the anisotropy parameter of the angular distribution, $P_2(\cos\theta)$ is a Legendre polynomial of degree 2, and θ is the angle between the axis of the electron-energy analyzer and the direction of polarization of the laser radiation.

Figure 2 shows the electron energy spectrum obtained at the angles $\theta=0°$ and 90°. The electronic spectrum consists of two peaks which correspond to the ground state $6s(^2S_{1/2})$ and two excited states $5d(^2D_{3/2,5/2})$ of Ba^+.

The angular distribution of the photoelectrons was investigated for electrons corresponding to the $6s$ state of the ion. The experimental curve of the intensity of the electronic signal versus the angle of rotation of the polarization plane of the radiation was approximated by the expression (2). This gave $\beta=0.74\pm0.1$.

A complete quantum-mechanical photoionization experiment requires measurement of the angular distribution to a high degree of accuracy with a low concentration of atoms in the beam. This condition must be satisfied to avoid possible screening of the electrons by the ions.[4] Difficulties with the recording system, which must meet exceedingly stringent requirements, still make it difficult in our case to increase the accuracy of the measurement of the parameter β.

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