Study of the lineshape of the autoionization resonance configuration $6p7p(^3P_1)$ of the barium atom

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Two-photon spectroscopy is used to study the lineshape of the autoionization resonance state $6p7p(^3P_1)$ of the barium atom. The autoionization resonance is excited via the intermediate states $6s6p(^1P_1)$ and $6s8p(^1P_1)$. The parameter q has been evaluated for both excitation channels by approximating the autoionization resonance lineshape with Fano's formula, yielding $q=11.1\pm1.0$ and 16.0 ± 2.0. As expected, the lifetime obtained for the autoionization state, 22.8 ± 1.0 cm^{-1}, is independent of the excitation method.

1. INTRODUCTION

We study two-step excitation of barium atoms from the ground state $6s^2(^1S_0)$ through the $6s6p(^1P_1)$ and $6s8p(^1P_1)$ intermediate states. The second excitation step transfers the atom into an autoionization state (AS) $6p7p(^3P_1)$. The product ions produced by AS decay are recorded by a time-of-flight mass spectrometer.

Camus *et al.*[1] were the first to study the $6p7p(^3P_1)$ AS. The state was identified as a result of calculations done in the Slater–Condon approximation. Experimental identification was carried out by Elizarov and Cherepkov[2] on the basis of selection rules for two-step excitation by polarized radiation.

2. EXPERIMENTAL SETUP

The experimental setup has been described in detail in Ref. 3, so here we briefly indicate the basic parameters. The experimental layout in shown in Fig. 1. The second harmonic from an LTI-405 Nd:YAG laser was used as the master oscillator for pumping two dye lasers. The specially constructed dye lasers had a linewidth of 2 cm^{-1}, frequency range 550–680 nm, and pulse energy 5 mJ. The wavelength of one dye laser was doubled in a KDP crystal. The radiation was linearly polarized using a Glan prism, which produced 99%-polarized radiation. A double Fresnel rhombus was used to rotate the polarization vector. The two counterpropagating laser beams (collinear to within 0.5 deg) were focused at their point of intersection, which was at the center of the barium-atom effusion beam. The latter was positioned between the two grids of the mass spectrometer, to which an electric field was applied. The expulsion pulse of the mass spectrometer had an peak value of 100 V, and the accelerating potential difference was 700 V. We used the surface ionization method to measure the intensity of the atomic effusion beam, which was maintained at a level no higher than 10^{10} cm^{-3}. The barium ions produced by photoionization were successively detected by two microchannel plates located 75 cm from the region where the radiation interacts with the atoms.

Along with the ion signal intensity, we measured the intensity of the second-step laser pulse, which ionizes the atoms, using a nanosecond photometer. The ion signal in each pulse is normalized to the second-step laser intensity. The value of the ion signal was averaged over 50 laser pulses. Figure 2 shows the energy-level diagram for barium atoms.

3. DISCUSSION OF THE RESULTS

We start with the excitation via the $6p6p(^1P_1)$ intermediate state that leads to the AS state $6p7p(^3P_1)$:

$$6s^2(^1S_0)+\lambda_1(553.5\ \text{nm})\rightarrow 6s6p(^1P_1),$$

$$6s6p(^1P_1)+\lambda_2(293.2\ \text{nm})\rightarrow 6p7p(^3P_1).$$

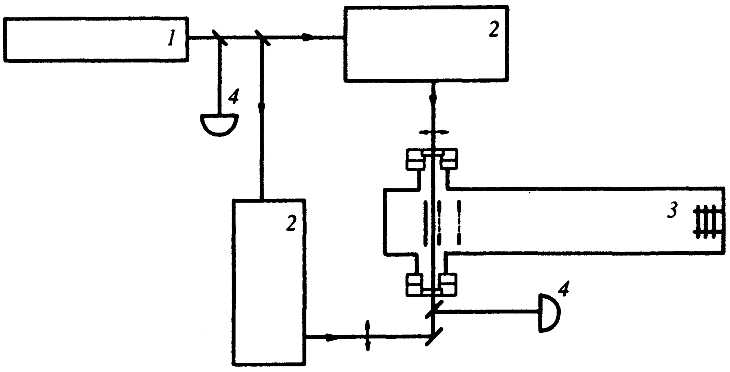

FIG. 1. Experimental layout: *1*—Nd:YAG laser; *2*—dye laser; *3*—time-of-flight mass spectrometer; *4*—photodiode.

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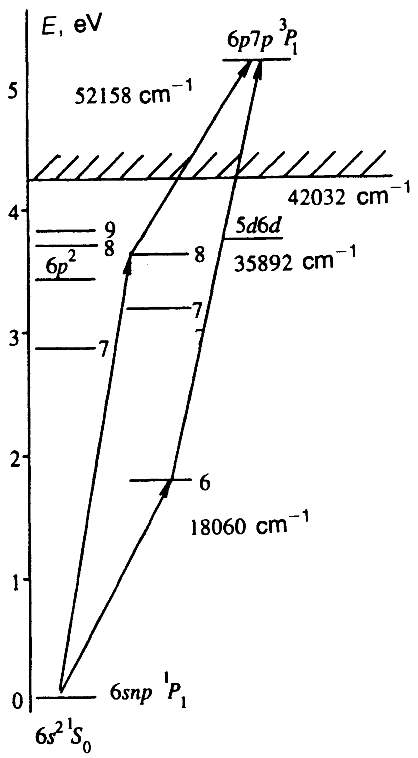

FIG. 2. Energy-level diagram of the barium atom and related transitions.

In this case the radiation of the resonance step (wavelength λ_1) is fixed and the radiation of wavelength λ_2 is scanned. There is no time lag between the two, which maximizes the photoionization signal.

At the above intensity, no two-photon ionization from the $6s6p$-state induced by λ_1-radiation was observed. The reason is that $\Omega \gg W^{(2)}$, where $W^{(2)}$ is the two-photon ionization probability. The photon flux in the λ_1-radiation was 10^{27} photons cm^{-2}s^{-1}. The Rabi frequency Ω for the transition between the $6s^2$ and $6s6p$ states is $\Omega=4.4\times10^{11}$ Hz. The probability of ionizing the excited state is approximately 10^9 Hz, so that after several picoseconds the $6s^2-6s6p$ transition is saturated and the $6s6p$ state can be assumed to be quasistationary.[4] The ion yield is a linear function of the intensity of the λ_2-radiation.

The AS lineshape can be described by Fano's formula[5]

$$\sigma(\varepsilon)=\sigma_a\frac{(q+\varepsilon)^2}{1+\varepsilon^2}+\sigma_b, \quad \varepsilon=\frac{E-E'}{0.5\Gamma}, \tag{1}$$

where E is the photon energy, E' is the AS energy, Γ is the autoionization resonance linewidth, σ_a is the photoionization cross section of the channels interacting with AS, and σ_b the photoionization cross section involving channels not interacting with AS.

Using the equations obtained in Ref. 6, we can represent these cross sections in the following manner:

$$\sigma_a\frac{(q+\varepsilon)^2}{1+\varepsilon^2}=\frac{4\pi^2\alpha\omega}{9}D_1^2, \quad \sigma_b=\frac{4\pi^2\alpha\omega}{9}\,\tfrac{3}{5}D_2^2,$$

where D_1^2 and D_2^2 are proportional to the sum of squares of the dipole matrix elements describing transitions to final states with spectral terms $J=1$ and $J=2$, respectively.

Figure 3 depicts the experimental autoionization resonance lineshape as a function of the second-step laser wavelength λ_2.

We approximated the experimental $\sigma(\varepsilon)$ curve by Eq. (1) and obtained

$$q=11.1\pm1.0.$$

The autoionization resonance linewidth Γ was found to be 22.8 ± 1 cm^{-1}.

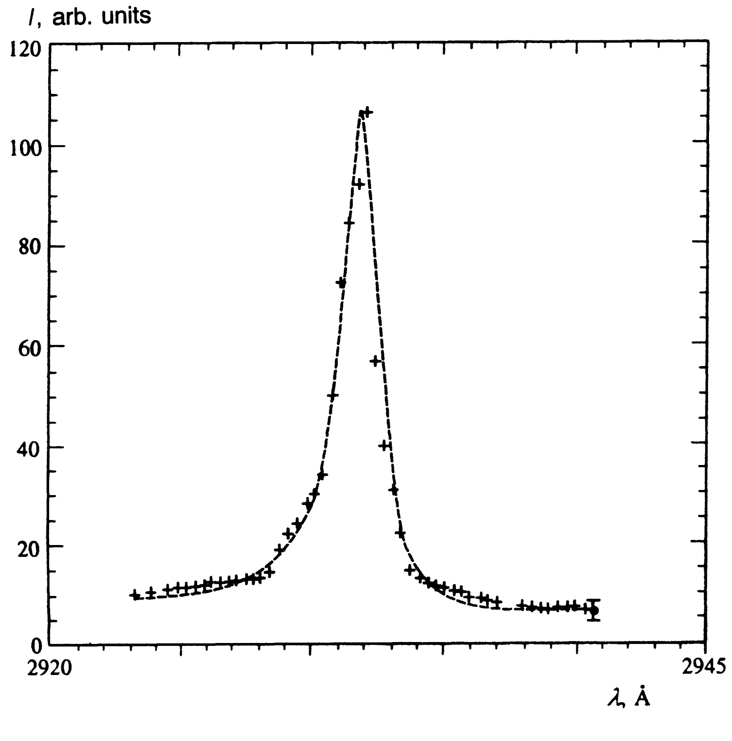

FIG. 3. Lineshape of the autoionization resonance state $6p7p(^3P_1)$ ($E=52\,158$ cm^{-1}) as a function of the second-step laser wavelength (intermediate resonance state $6s6p(^1P_1)$); the lasers are linearly polarized in mutually perpendicular planes [+]. The dot-dash curve is the result of approximation by Eq. (1), with $q=11.1\pm1.0$.

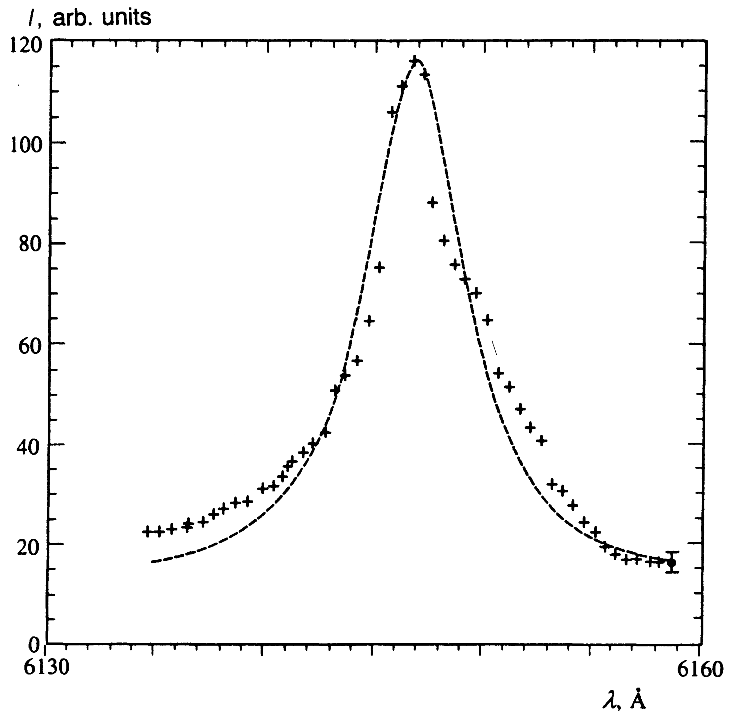

FIG. 4. Lineshape of the autoionization resonance state $6p7p(^3P_1)$ (E=52 158 cm^{-1}) as a function of the second-step laser wavelength (intermediate resonance state $6s8p(^1P_1)$); the lasers are linearly polarized in mutually perpendicular planes [+]. The dot-dash curve is the result of approximation by Eq. (1), with $q=16.0\pm2.0$.

The excitation via the $6s8p(^1P_1)$ intermediate state that leads to the AS state $6p7p(^3P_1)$ is

$$6s^2(^1S_0)+\lambda_1(278.6\ \text{nm})\rightarrow 6s8p(^1P_1),$$

$$6s8p(^1P_1)+\lambda_2(614.7\ \text{nm})\rightarrow 6p7p(^3P_1).$$

The results of measuring the AS lineshape with excitation through the intermediate state $6s8p(^1P_1)$ are illustrated by Fig. 4. Note that in addition to ionization decay of AS, there is an ionization channel out of the $6s8p(^1P_1)$ state via emission of λ^1 radiation. However, this channel is not particularly influential, since the intensity is ten times lower at λ_1 than at λ_2; furthermore, the AS ionization cross section exceeds the ionization cross section without AS excitation by approximately two orders of magnitude.

In addition to the experimental AS lineshape, Fig. 4 shows the curve obtained by fitting with Fano's formula (1), which yielded a value $q=16.0\pm2.0$. The AS linewidth Γ for this excitation channel is 21 ± 2 cm^{-1}. As expected, to within the experimental errors, the value of Γ is the same for the two AS excitation channels.

4. CONCLUSION

The lineshape of the autoionization resonance state $6p7p(^3P_1)$ of the barium atom was studied for the first time experimentally. We have found the values of the parameters q and Γ for two excitation channels through different intermediate states, $6s6p$ and $6s8p$. The resonance linewidths for these excitation channels are the same, and the values of q turn out to be different. In particular, the data can be used to advance the theory of the quantum-defect method, which has been successfully used to describe AS spectra at lower energies, where only one ionization channel is open.[7]

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