Excitation of high-lying state of the sodium atom by dye-laser radiation and their autoionization in an electric field

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Ionization of Na atoms by an electric field was realized for the first time with selected two-stage excitation by emission from pulsed dye lasers. The cross section for the photoionization of Na atoms via the autoionization state $15d$ in an electric field 12 kV/cm amounts to 0.7×10^{-14} cm^2.

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1. To separate isotopes and isomers, and to obtain ultrapure substances, interest attaches to the method of selective ionization of atoms by laser radiation. A two-stage scheme for the ionization of atoms by an optical field was first proposed and realized (the Rb atom) in[1]. Recently, a similar scheme was used to separate uranium isotopes.[2] A significant shortcoming of this method is the small photoionization cross section σ_i of the atoms (a typical value is $\sigma_i \approx 10^{-17}$–$10^{-19}$ cm^2).[3]

To increase the ionization cross section, it was proposed in[4] to use the phenomenon of autoionization decay of states near the ionization limit of the atom under the influence of an external constant electric field. In this study, this ionization method was realized experimentally, using as an example the Na atom excited by two tunable lasers into states close to the ionization limit.

2. The experimental setup is shown in Fig. 1. A nitrogen laser with a transverse discharge (1) (pulse repetition frequency 10 Hz, pulse duration $\tau = 9$ nsec) excited simultaneously two tunable dye lasers. For resonant excitation of the $3^2P_{1/2}$ state of Na, the laser (3) (rhodamine-6G) was tuned to $\lambda_1 = 5895.9$ Å. The width of the lasing spectrum was 0.3 Å, the radiation power was 0.5 kW, which certainly ensured saturation of the $3^2S_{1/2} \rightarrow 3^2P_{1/2}$ transition. The wavelength of laser (4) (POPOP solution in toluene + KOH) was tuned in the range 4210–4140 Å, so that it was possible to excite resonantly the high-lying S and D states of Na with quantum numbers n ranging from 13 to 18. The width of the laser emission spectrum was 0.1 Å, and the energy in the pulse was 0.5×10^{-7} J. The light beams were directed to a cell with Na vapor, where they crossed in the region between the electrodes. The elec-

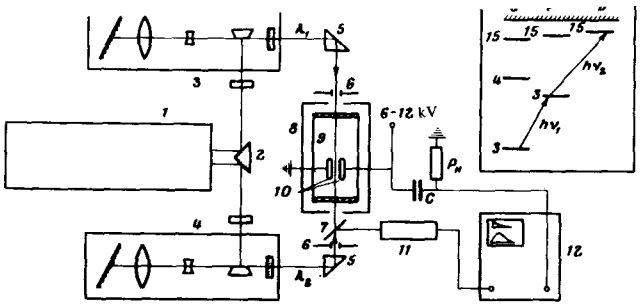

FIG. 1. Experimental setup: 1—nitrogen laser, 2—beam splitter, 3, 4—tunable dye lasers, 5—rotating prisms, 6—diaphragms, 7—beam-splitting plate, 8—heater, 9—cell with sodium vapor, 10—electrodes, 11—photomultiplier, 12—oscilloscope.

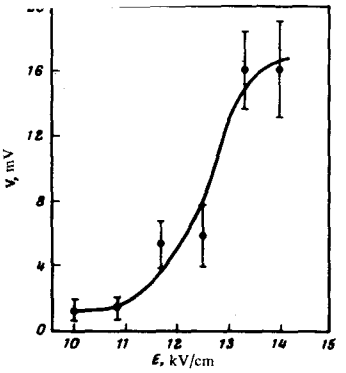

FIG. 3. Dependence of the two-stage ionization signal on the electric field following resonant excitation of the $15^2D_{3/2}$ state of Na.

trodes (20 mm diameter) were located 6 mm apart. The cell temperature was $t = 175 \pm 3$ °C. The dc voltage on the electrodes ranged from 6 to 12 kV. The photoionization signal and the radiation pulse of frequency λ_2 were registered by a two-beam oscilloscope. The laser of wavelength λ_1 saturated the absorption of the resonant transition of Na over the entire length of the cell (≈ 10 cm), a fact visually monitored by the produced fluorescence.

3. Figure 2 shows the dependence of the ionization signal (in millivolts) on the wavelength λ_2 in the vicinity of the $3^2P_{1/2} \rightarrow 15^2D_{3/2}$ line at different electric field intensities (the $15d$ level lies 488 cm^{-1} below the ionization limit). The signal constitutes a difference of two signals, a resonant signal observed when the beams of the first and second stages act simultaneously, and a nonresonant signal produced by the action of only the second stage. Radiation of only the first stage produced no photocurrent. The resonant signal vanished also when the wavelength λ_1 was detuned from exact resonance. The figure shows clearly the resonant character of the two-stage ionization as a function of the wavelength λ_2, and the dependence of the ionization signal on the electric field intensity. Resonant photoionization of this kind was observed upon excitation of any of the S and D states of Na with a principal quantum number that ranges from $n = 12$ to $n = 18$. The dependence of the ionization signal on the electric field intensity in the case of exact tuning of λ_2 to resonance is shown in Fig. 3 (the nonresonant part was subtracted from the ionization signal). An abrupt increase of the photocurrent upon excitation of the $15d$ state occurs in a field $E = 12$ kV/cm. The lifetime of the $15d$ state is $\tau_{15d} \approx 10^{-9}$ sec. Therefore a noticeable autoionization signal should occur in fields at which the autoionization probability is $W_{aut} \approx 1/\tau_{15d}$. A theoretical calculation (formula (4) of[4]) yields a field value $E = 11$ kV/cm for a principal quantum number $n = 15$ (parabolic quantum number $n_2 = 7$). By monitoring all the parameters of the experiment it is also possible to compare the photoionization signal with the calculated value. The amplitude of the ionization signal in the experiment was $V = Q/C$, where Q is the charge produced in the course of the ionization and C is the capacitance of the measuring circuit (240×10^{-12} F). The maximum charge Q at $W(E) \gtrsim 1/\tau_{15d}$ is determined by the number of particles excited into the state $15d$. It is equal to $N_i = n\sigma_2 \mathcal{E}(\lambda_2)\Delta\omega_D/\Delta\omega_L$, where n_1 is the number of atoms in the $3^2P_{1/2}$ state, σ_2 is the cross section for the excitation of the $15^2D_{3/2}$ state from the $3^2P_{1/2}$ state, $\mathcal{E}(\lambda_2)$ is the energy density of the second-stage laser, $\Delta\omega_D$ is the Doppler width of the line, and $\Delta\omega_L$ is the laser line width. In the experiment, $n_1 = 3 \times 10^9$ (particle concentration 10^{11} cm^{-3}, volume 6×10^{-2} cm^3), and $\mathcal{E}(\lambda_2) = 3.5 \times 10^{12}$ photons/cm^2 (energy 5×10^{-8} J, area 3.14×10^{-2} cm^2). The cross section σ_2 was determined by extrapolating the dependence of the transition probability A_{3p-nd} on n, which is known up to $n = 13$,[15] and which is equal to 7.3×10^{-15} cm^2. We thus obtain for the signal the value $V = 5 \times 10^{-3}$ V. The ionization signal observed at $E = 12$ kV/cm (Fig. 3) was 4×10^{-3} V.

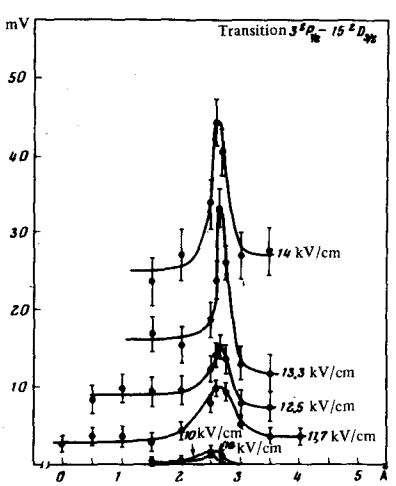

FIG. 2. Dependence of the signal of two-stage ionization on the wavelength of the second-stage laser and on the electric field intensity following excitation of the $15d$ level of Na.

4. Thus, the maximum cross section for the photoionization of an atom in an electric field is determined by the cross section σ_2 of the $3p \rightarrow 15d$ transition ($\sigma_2 = 0.7 \times 10^{-14}$ cm^2 for Na). In an electric field $E = 12$ kV/cm, the Na photoionization cross section is increased by a factor $\sigma_2/\sigma_i \approx 10^5$ in comparison with the cross

section of the nonresonant (in the second state) two-stage ionization ($\sigma_i = 10^{-19}$ cm^2).[6] This greatly extends the possibilities of using the method of selective photoionization of atoms by laser radiation.

[1]R. V. Ambartsumyan, V. P. Kalinin, and V. S. Letokhov, ZhETF Pis. Red. **13**, 305 (1971) [JETP Lett. **13**, 217 (1971)].

[2]B. Snavely, Report on VIII Intern. Quant. Electr. Conf. July, 1974, San Francisco.

[3]I. I. Sobel'man, Vvedenie v teoriyu atomnykh spektrov (Introduction to the Theory of Atomic Spectra), Moscow (1963).

[4]L. N. Ivanov and V. S. Letokhov, Kvantovaya ėlektronika **2**, 585 (1975) [Sov. J. Quant. Electr. **5**, No. 3 (1975)].

[5]W. L. Wiese, M. W. Smith, and B. M. Miller, Atomic transition Probabilities (Sodium through calcium), NSRDS-NBS **2**, 22 (1969).

[6]U. Fano and J. Cooper, Spectral Distribution of Oscillator Strengths in Atoms (Russ. transl.), M., 1972.