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Ionization of high-lying states of the sodium atom by a pulsed electric field

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The critical field intensities are obtained for the S and D states with $n = 12$–19 of a sodium atom excited by radiation from a pulsed dye laser with continuously tunable lasing frequency. The experimentally measured critical intensity agrees well with the calculated one and is proportional to $(n^*)^{-4}$, where n^* is the effective principal quantum number. It is shown that in a field with an intensity higher than critical, the highly-excited atoms are ionized with an ion yield equal to unity. The effective cross section of the atom-ionization process is equal in this case to the cross section for the excitation of the high-lying states.

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1. INTRODUCTION. FORMULATION OF PROBLEM

Much attention is being paid recently to selective stepwise photoionization of atoms by laser radiation. This method is promising for the solution of such problems as isotope separation, nuclear isomers, production of ultrapure substances, etc., since it is universal, i.e., it is applicable practically to all elements and is effective.

The gist of the method consists in the following. The atoms of the chosen species are selectively excited into the intermediate state by narrow-band laser radiation. Additional laser radiation is then used to ionize the excited atoms. This ionization method and its use for separation of atoms was first proposed in[1] and realized in[2] with Rb as an example. This method was then used to carry selective photoionization of the Ca^{40} isotope[3] and a weighable amount of uranium enriched with U^{235} was obtained[4,1)]. In,[7] using a tunable dye laser or by using the emission of a ruby laser and its second harmonic for the ionization, photoionization of rubidium atoms was effected with an ion yield close to unity. The resultant ions and electrons made up a plasma with a maximum charge density $\approx 10^{13}$ cm^{-3}.

Despite the high selectivity and efficiency of the method in individual experiments, its extensive use is difficult. The reason is that, owing to the smallness of the cross section σ_{ion} for the ionization of the atom (σ_{ion}

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$\approx 10^{-17}$–10^{-19} cm^2), the photoionization rate ($W_{ion} = \sigma I$) becomes comparable with the rate of the radiative decay only in laser fields with intensity on the order of several MW/cm^2. As a result, to extract the maximum number of selected atoms from a stream moving with thermal velocity, it is necessary to use lasers with an average radiation power on the order of several kilowatts. Therefore a search for ways of increasing the cross section of the ionization of the atoms from the excited state is quite pressing.

To decrease the atom ionization cross section, it was proposed[8] to use autoionization decay of a highly excited atom in an external electric field. The gist of the process is the following. Under the influence of the electric field, a fraction of the high-lying states (Figs. 1a, b) goes over into the continuous spectrum, and the remainder is in the auto-ionization state as a result of below-the-barrier transition. The closer the state to the ionization limits, the larger the rate of the photoionization decay, and this rate increases rapidly with increasing principal quantum number n and with increasing electric field intensity E. The transition of the discrete states into the continuum takes place in a field of intensity

$$E > E_{cr} = 1/16n^{*4} \text{ a.u.} \quad (1)$$

(1 a. u. = 5×10^9 V/cm, n^* is effective principal quantum number).

The electric field intensity can be chosen such that the rate of the autoionization becomes comparable with or exceeds the rate of the radiative decay. The probability W_{aut} of ionization of an atom from a highly-excited state approaches unity in this case, and the cross section for ionization from a selectively excited state, if we define it as $\sigma_{ion} = \sigma_2 W_{aut}$, becomes equal to the cross section σ_2 of the excitation of a high-lying state (usually $\sigma_2 = 10^{-12}$–10^{-15} cm^2). Thus, the use of an electric field makes it possible to increase the cross section of the photoionization of the atom by four or five orders.

The proposed ionization method was realized in experiment with the sodium atom as an example.[10] The ionization was effected in a constant electric field. It was shown that the phenomenon has a strongly pronounced threshold. For states excited by a laser with wavelength 4167 Å, the threshold intensity was 12 kV/cm.

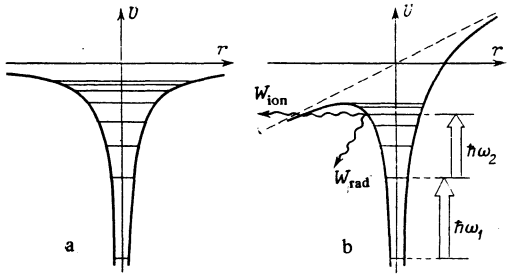

FIG. 1. Energy of an electron in the Coulomb field of a nucleus: a) in the unperturbed state, b) when an external constant electric field is applied.

It should be noted that at field intensities when the autoionization becomes noticable, a strong Stark effect also takes place. As a result, the high-lying levels split into a large number of components, the levels with different n intersect,[11] and this leads to an uncertainty in the quantum numbers of the excited state. A preferable scheme is one in which the electric field is turned on after the laser pulses. In this case the excited states are those whose position is known with high accuracy. Ionization of sodium atoms ($n = 26$–37), zenon atoms ($n = 24$–28, f states), and uranium atoms ($n = 40$–50) by a pulsed electric field was investigated by a number of workers.[12–14]

Inasmuch as on going to a high-lying state the absorption line width is of the order of the Doppler width, effective excitation of these states calls for a narrow-band smoothly tunable laser. The generation efficiency of such lasers is usually several percent. Therefore ionization via high-lying states is favored over ionization directly from a selectively excited state in the case when the efficiency η_{nar} of a laser with line width comparable with the Doppler width and the efficiency η_{br} of a broadband laser for direct ionization via the continuum satisfy the condition

$$\eta_{nar}/\eta_{br} \geqslant \sigma_{ion}/\sigma_2 W. \quad (2)$$

The excitation cross section σ_2 is proportional to n^{-3}. It is therefore obvious that the maximum gain when using high-lying states will occur at not too large values of n. On the other hand, the critical field, i.e., the field at which the ion yield is close to unity, increases sharply with decreasing n. The optimal case is one in which the intensity of the electric field is of the order of 10 kV/cm. In such a field the states with $n \approx 15$ are easily ionized.

The purpose of the present study is to investigate the process of ionization of sodium atoms from high-lying states $n = 12$–19 in a pulsed electric field.

2. EXPERIMENTAL PROCEDURE AND DESCRIPTION OF SETUP

In this study, just as in the study of Ducas *et al.*,[12] to eliminate collision ionization of highly excited atoms we used an atom beam. The sodium atoms in the beam were excited to high-lying states by radiation of two pulsed dye lasers with smoothly tunable lasing frequency. The energy level scheme of the sodium atom and the employed transitions are shown in Fig. 2. One of the lasers ($\lambda_1 = 5890$ Å) excited the resonant $3^2P_{3/2}$ state. The wavelengths of the second laser were varied in the range 4250–4140 Å, and this made it possible to excite S and D states with principal quantum number $n = 12$–19 from the $3^2P_{3/2}$ state. The light beams intersected the atomic beam in the region between the electrodes. A rectangular electric pulse was applied to the electrode. The produced ions were drawn up from the ionization region through a slit in one of the electrodes and were registered with a secondary electron multiplier.

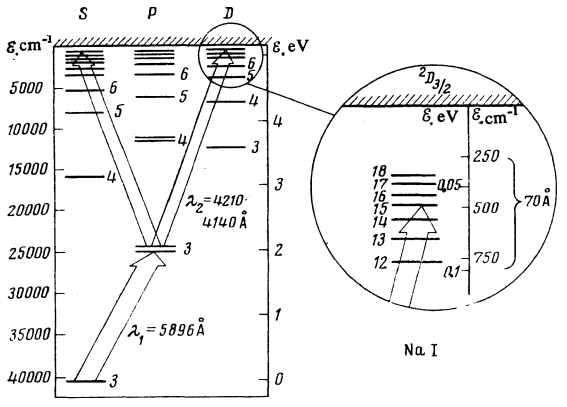

FIG. 2. Energy level scheme of sodium atoms, and employed transitions.

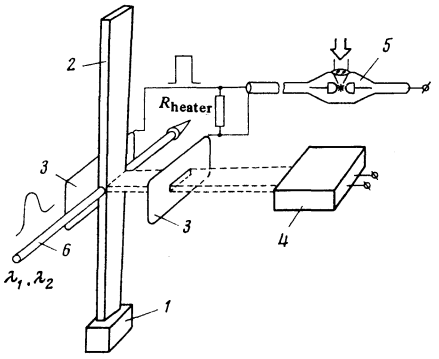

FIG. 4. Relative placement of the light beams, the atomic beam, the electrodes, and the ion beam. 1—oven with collimating diaphragm to produce the atom beam, 2—sodium-atom beam, 3—electrodes, 4—secondary electron multiplier, 5—matched optically ignited discharge gap, 6—laser light beams.

The experimental setup is shown in Fig. 3. A nitrogen laser with a transverse discharge 1 (pulse power 100 kW, duration ≈10 nsec, repetition frequency 10 Hz) excited simultaneously two dye lasers. The lasers consisted of the following elements: cells containing the dye (without circulation), mirrors with reflection coefficients 4%, diffraction gratings—1200 lines/mm, first order; telescope with 25-fold magnification to decrease the divergence of the light beam incident on the grating. The radiation of the nitrogen laser was focused into the cell with the dye by a cylindrical quartz lens of focal length 12 cm.

The medium in the laser tube of the first excitation stage was a solution of rhodamine 6Zh in ethanol. The width of the generation spectrum was $\Delta\lambda_1 = 0.3$ Å, the output energy $E_1 = 8$–10 μJ, the generation pulse duration $\tau_1 = 7$ nsec. These laser parameters were sure to produce saturation of the absorption the $3^2S_{1/2} - 3^2P_{3/2}$ transition. The second laser 3, using a POPOP solution in toluol, had the following parameters: $E_2 = 6$–7 μJ, $\tau_2 = 8$ nsec, and $\Delta\lambda_2 = 0.2$ Å.

The laser beams were diverted with semitransparent mirrors to the working vacuum chamber 4 and to an additional cell with sodium vapor 5. With the aid of this cell, which was placed in a heater, we monitored the frequency and energy of the lasers by observing the luminescence signal from the high-lying state, which was registered by photomultiplier 6.

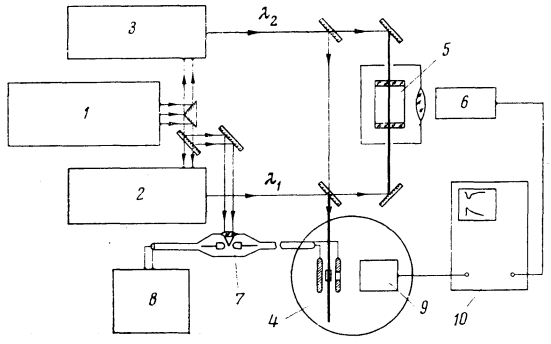

FIG. 3. Experimental setup: 1—pulsed nitrogen laser, 2—rhodamine 6Zh laser—first excitation state, 3—POPOP dye laser—second excitation stage, 4—vacuum chamber, 5—cell with sodium vapor placed in a heater, 6—photomultiplier, 7—matched optically-ignited discharge gap, 8—high-voltage source, 9—secondary electron multiplier, 10—two-beam oscilloscope.

The atomic beam was formed as a result of effusion of the sodium vapor through a slit from an oven heated to 220 °C (approximate vapor pressure 10^{-3} Torr). The beam was collimated by a diaphragm in such a way that in the region between the electrodes it had a cross section of 2×6 mm. The concentration of the atoms in the beam was 10^8–10^9 cm^{-3}. The residual pressure in the chamber did not exceed 2×10^{-6} Torr.

A voltage pulse was produced on the electrodes when a section of coaxial cable was discharged through a matched optically-ignited discharge gap into the transmission line 7. To trigger the pulse, part of the radiation of the nitrogen laser was diverted and focused on one of the electrodes. This system made it possible to apply to a matched load a single voltage pulse of rectangular shape with duration 8 nsec and amplitude from 1 to 30 kV, synchronized with high accuracy with the laser pulses. The delay of the high-voltage pulse at the output of the gap relative to the triggering pulse of the N_2 laser did not exceed 5 nsec in a discharge-gap pressure range from 6 to 10 atm. The length of the transmission line was chosen such that the electric pulse was produced on the electrodes in the region of the interaction of the atomic and light beams at an instant 35 nsec following the laser pulses.

The ions produced as a result of autoionization of the highly-excited atoms (Fig. 4) were drawn out through a slit in the electrode with zero potential by the electric field of the cathode of a secondary electron multiplier (SEM) whose potential was −4 kV. To decrease the dependence of the output signal of the SEM on the energy of the registered ions, the duration of the pulse shaped by the discharge gap was chosen equal to 8 nsec. The maximum energy acquired by the ions in the region between the electrodes did not exceed 0.3 keV, and was equal to 4 keV in the SEM field. Signals from the SEM and the multiplier, which controls the dependence of the highlying states, were registered with a two-beam oscilloscope 10.

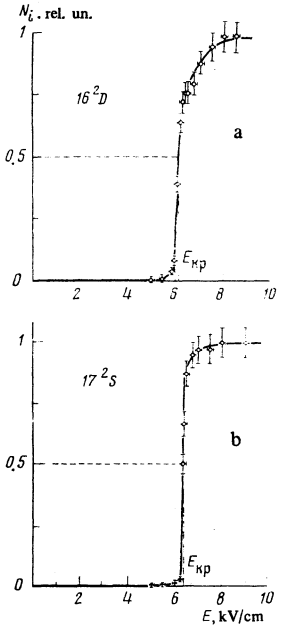

FIG. 5. Dependence of the ion yield on the electric field intensity: a—excitation of the $16\,^2D$ level; b—excitation of the $17\,^2S$ level.

3. RESULTS OF EXPERIMENT

Figure 5 shows the plots of the yield of ions against the electric field intensity following excitation of the 17^2S and 16^2D states of the sodium atom. The plots have clearly pronounced thresholds. In the absence of a field, the registered ion signal is selective in the frequency of the lasers of both excitation steps. When the field intensity is increased to the critical value, the signal increases slightly. When the critical value is reached, the signal increases abruptly by three–four orders of magnitude. Further increase of the electric field intensity leads to saturation of the ion signal. The D state is characterized by a smoother dependence of the ion signal on the electric field intensity than the S state.

The selective ion signal observed in the absence of an electric field can arise both following ionization of highly excited sodium atoms by laser radiation and following ionization of the atoms as a result of their collisions. The small increase of the signal with increasing field intensity is apparently due only to the more complete extraction of the ions from the interelectrode space, since the probability of autoionization in the electric field in this region of field intensities is exceedingly low. The abrupt increase of the signal when the field intensity goes through the critical value is due to the very strong dependence of the ionization probability on the electric field intensity. The signal saturates upon ionization of practically all the atoms excited to a given state within a time equal to the duration of the electric-field pulse. Since the D state splits in the electric field into several sublevels with different values of the critical field, the ion signal reaches saturation more smoothly than for the S state, which undergoes only a shift.[11]

Analogous threshold dependences were obtained for the S states $n = 13$–19 and D states $n = 12$–18. From the obtained curves it is possible to determine the critical values of the electric field intensity. The critical values were assumed to be field intensities at which half the highly-excited atoms became ionized; this corresponds to an ion signal half as strong as in saturation. We present below, for all the investigated states, the critical values of the electric field intensity:

n:	19	18	17	16	15	14	13	12
^2S n^*:	17.65	16.65	15.65	14.65	13.65	12.65	11.65	
^2S E_{cr}, kV/cm	4.1	4.8	6.4	8.3	10.8	15.4	20.6	
n^*:		18	17	16	15	14	13	12
^2D E_{cr}, kV/cm		3.9	4.7	6.2	8.2	10.6	14.2	20.2

The method of ionization of highly excited atoms by an electric field can be used to measure the number of these atoms.[11] Compared with the fluorescence procedure, this method has the following advantages. The ion yield is practically equal to the number of highly excited atoms, whereas the number of photons emitted as a result of radiative decay in the registered-frequency band is small. This is determined both by the large number of intermediate states and by the small solid angle of the recording apparatus, as well as by the short time that the atom is situated in the light beam in comparison with the lifetime of the high-lying states, which is proportional to n^3. By way of an example of such a method of detection of highly excited atoms, Fig. 6 shows the resonant dependence of the yield of the ions on the radiation frequency of the laser of the second stage, obtained when the frequency of this laser was tuned and the number of produced highly excited atoms was detected by measuring the yield of the ions under the influence of an electric field stronger than the critical field. The width of this curve is determined by the width of the laser emission line. We used the transition $3^2P_{3/2} \rightarrow 17^2S_{1/2}$ at a field intensity $E = 9$ kV/cm.

4. COMPARISON WITH THE THEORY

The dependence of the critical electric field intensity on the effective quantum number n^* can be described theoretically by two approaches. A classical analysis of the ionization of an atom by an electric field[9] yielded formula (1), which can be derived from the condition that the level energy be equal to the value of the potential at the maximum (Fig. 1b). A shortcoming of this approach is the neglect of the below-barrier transition of the electron and the shift of the level due to the Stark effect. This problem was solved quantum-mechanically

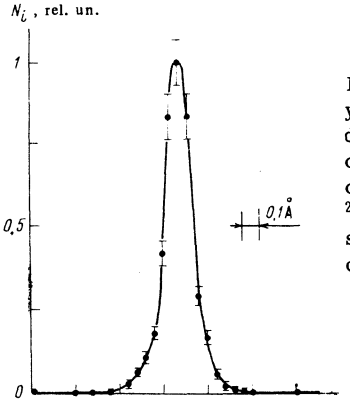

FIG. 6. Dependence of the ion yield on the variation of the radiation wavelength of the second-stage laser in the vicinity of the transition $3\,^2P_{3/2} \rightarrow 17\,^2S_{1/2}$ at an electric field intensity $E = 9$ kV/cm exceeding the critical value.

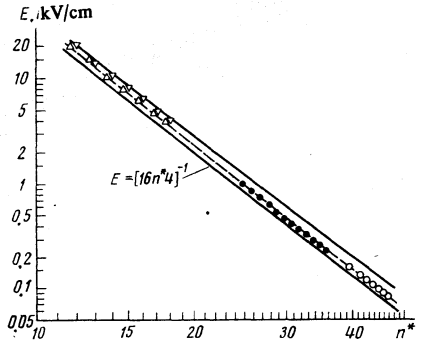

FIG. 7. Dependence of the critical intensity of the electric field E_{cr} on the effective principal quantum number n^* of the excited level: △, ▽—results of present paper for the S and D states, respectively; ●—from[12], ○—from[14].

with allowance for the tunneling of the electron in[8]. The following expression was obtained for the rate of autoionization in first-order perturbation theory:

$$W=\frac{1}{n^2}\left(\frac{4}{En^3}\right)^{1+|m|+2n_2}\frac{1}{n_2!\,(n_2+|m|)!}\exp\left\{-\frac{2}{3En^3}+3(n-2n_2-|m|-1)\right\} \quad (3)$$

where E is the intensity of the electric field in atomic units, n is the principal quantum number, n_2 is the parabolic quantum number, and m is the magnetic quantum number. In this formula, account was taken of both the linear Stark effect and the below-barrier transition of the electron.

Figure 7 shows the dependence of the critical intensity of the electric field on the effective principal quantum number n^*, calculated from formula (1) (lower line) and calculated from formula (3) (for a pulsed electric field with a pulse duration τ_p equal to the lifetime of the corresponding high-lying state τ_{rad} (upper line). For $\tau_p = 8$ nsec $\ll \tau_{rad}$ (this corresponds to the conditions of our experiment), the corresponding theoretical dependence lies 6% above the upper curve, i.e., agrees better with experiment than the simple formula (1). The same figure shows the value of the critical intensity, taken from the data presented above, and also the results obtained in[12] (n= 26–37, sodium atom) and[14] (n= 40–50, uranium atom). The experimental values of all these studies differ somewhat from both calculated relations. The discrepancy between the experimental and theoretical results exceeds the accuracy limits of the experiment and amounts to 15% on the average.

The parallelism between the classical relation $E_{cr} = 1/16n^{*4}$ and the approximation curve (dashed line) constructed from the experimental results of[12,14] and of the present study is clearly seen. Thus, the critical electric field intensity measured in the experiment is proportional to $(n^*)^{-4}$, and the approximation curve can be expressed by the relation

$$E_{cr}^{(exp)}=0.07\,(n^*)^{-4}. \quad (4)$$

Thus, experiment has shown that the critical electric field intensity at which half the highly-excited atoms is ionized is described by relation (4). The difference between the experimental critical intensities and the theoretical ones calculated by formula (3) does not exceed 15%.

In an electric field stronger than critical, the highly-excited atoms are ionized with an ion yield equal to unity. The effective cross section of the entire ionization process is determined here by the cross sections for the resonant excitation of the low-lying state. In a moderate electric field $E \lesssim 20$ kV/cm, the ionization cross section can be raised to a value 10^{-13} cm^2.

The proposed method of ionizing excited atoms solves in practice the problem of choosing the optimal system of selective stepwise photoionization of atoms when low-power lasers (on the order of 0.1–1 W) and laboratory installations of moderate dimensions (lengths on the order of 0.1–1 m) are used. The method can be employed for effective separation of isotopes, nuclear isomers, and obtaining ultrapure substances. The proposed method is therefore of undisputed practical interest.

[1)]A brief survey of the applications of the method of selective ionization of atoms for isotope separation is contained in[5]. A more complete review of the method and its application is given in[6].

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