Laser photoionization detection of individual sodium atoms via Rydberg states

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Individual atoms have been registered for the first time by using the method whereby the atom is ionized by an electric field from high-lying (Rydberg) states. Sodium atoms excited to the $13d$ state by radiation from pulsed dye lasers were ionized by an electric-field pulse. 3–5 sodium atoms from the atomic beam were registered in the observation region.

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1. The method of selective stepwise ionization of atoms by laser radiation, first realized in[1], can be used not only to separate isotopes but also to detect atoms, inasmuch as at sufficiently high energy density of the laser pulses the photoionization quantum yield is close to unity.[2] This possibility was pointed out in[1–3]. The first successful experiment aimed at detecting individual atoms by selective photoionization was made in[6].

A shortcoming of the method of two-step ionization of atoms by laser radiation is the relatively low photoionization cross section compared with the resonant-excitation cross section. This imposes very stringent requirements on the energetics of the ionizing laser pulse, particularly if one registers atoms that cross the laser beam with thermal velocity, when a high pulse repetition frequency is necessary to intercept all the atoms.

The laser-pulse energy necessary to ionize the atom can be decreased by several orders if the atom is ionized from a high-lying state by an electric field, as was proposed in[7] and realized in[8].

In the present study we have used, for the first time ever, a method of ionizing high-excited atoms by an electric field to register individual sodium atoms.

2. The sodium atoms in the beam (Fig. 1a) were excited in two steps into a high-lying state by pulsed tunable-frequency dye lasers (spectral width $\Delta\nu \approx 1$ cm^{-1}). The lasers were pumped simultaneously with a nitrogen laser at a pulse repetition frequency 10 Hz. The first laser transferred the sodium atoms from the ground state to the $3^2P_{3/2}$ state, while the second laser from the $3^2P_{3/2}$ state to the Rydberg state $13d$ (Fig. 1b). The light beams crossed the atom beam in the region between the electrodes, and the volume of the intersection zone of all the beams was $\pi(0.5)^2 \times 6$ mm^3. At 20 nsec following the laser pulses, a square-wave voltage pulse was applied to the electrodes. The produced ions were drawn out of the ionization volume through a slit in one of the

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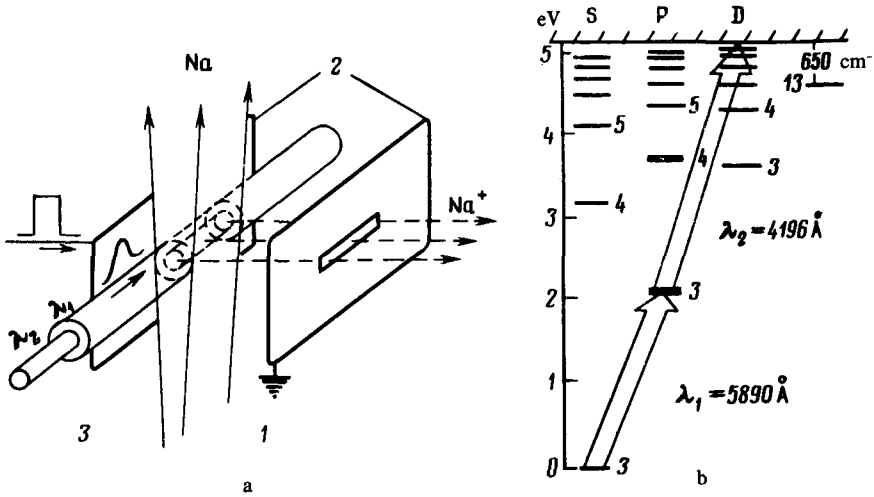

FIG. 1. a) Relative placement of the atomic and light beams: 1—sodium-atom beam, 2—electrodes, 3—laser light beams. b) Scheme of the employed Na atom transitions.

electrodes and were registered by a secondary-electron multiplier (SEM). The experimental setup is described in greater detail in[9]. The principal quantum number of the Rydberg state was chosen such that the cross section for the excitation of this state was as large as possible, and the electric field intensity at which the ion yield is close to unity was easily attainable under laboratory conditions. The state 13*d* satisfies these requirements.

3. Figure 2(a) shows the dependence of the ion yield on the intensity of the pulsed electric field when the sodium atoms are excited into the 13*d* state. This dependence has a clearly pronounced threshold. The abrupt increase of the signal on going through the critical value of the electric field is due to the very strong dependence of the ionization probability on the electric field intensity. Saturation of the ion signal sets in when all the atoms excited to the 13*d* state are ionized during the electric-field pulse. By using this dependence, the working value of the electric field intensity was chosen to be 17 kV/cm.

Figures 2(b) and 2(c) show plots of the ion yield against the energy density of the laser pulses in the first and second stages of excitation, at an electric field intensity 17 kV/cm. The saturation energy densities of the first and second transitions, determined from these relations, are $E^{(1)}_{sat}=3\times10^{-7}$ J/cm^2 and $E^{(2)}_{sat}=4\times10^{-5}$ J/cm^2, respectively. The results agree well with the calculations for the first stage, and are of the same order of magnitude for the second stage. The atoms were detected at a laser-pulse energy 20 μJ. In this case, approximately 2/5 of all the particles interacting with the laser field are excited into the 13*d* state.

Figure 3 shows the dependence of the yield of the ions on the temperature of the sodium oven. It shows also the dependence, calculated in standard fashion, of the

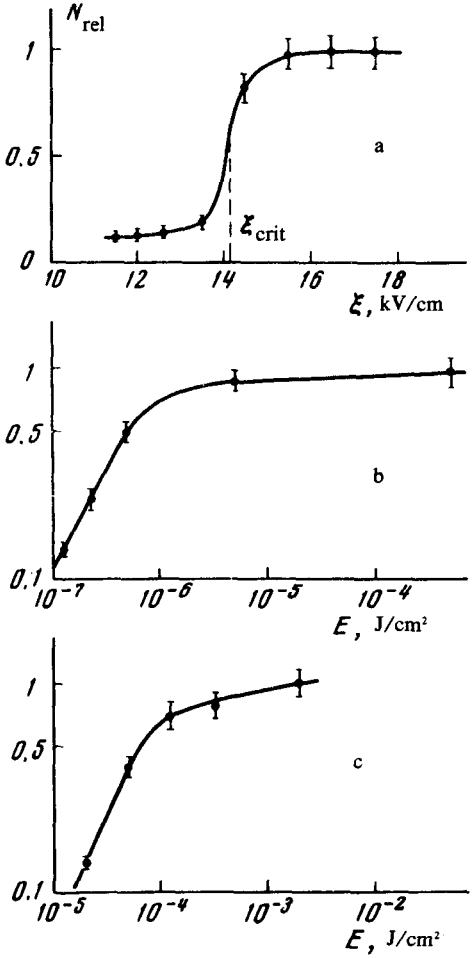

FIG. 2. a) Dependence of the ion yield on the electric field intensity when Na atoms are excited into the 13*d* state. b) Dependence of the ion yield on the energy density of the laser pulse of the first excitation stage, electric field intensity $E=17$ kV/cm. c) Dependence of the ion yield on the energy density of the laser pulse of the second excitation stage, $E=17$ kV/cm.

number of beam atoms interacting with the laser radiation, on the temperature (dashed curve). At an oven temperature $t_n > 100$°C, these curves are parallel. Therefore the experimental dependence was normalized in such a way that it coincided with the value calculated at $t_n \geqslant 100$°C. With decreasing temperature, the detected number of atoms differs from the calculated one. The apparent reason is that the atomic vapor in the oven at this temperature is not saturated. The minimal signal was registered at an oven temperature 55 °C and was equal to 3–5 atoms. The theoretical relation yields at this temperature a maximum of 9 atoms in an excited volume $V=5\times10^{-3}$ cm³. Comparison of the amplitude of the minimal signal with the amplitude of the dark "single-electron" pulses of the SEM attests to registration of 1 or 2 sodium atoms, in good agreeement with the experimental value. The additional instability of the ion signal, observed at the minimal values of the oven temperature, is due to fluctuations

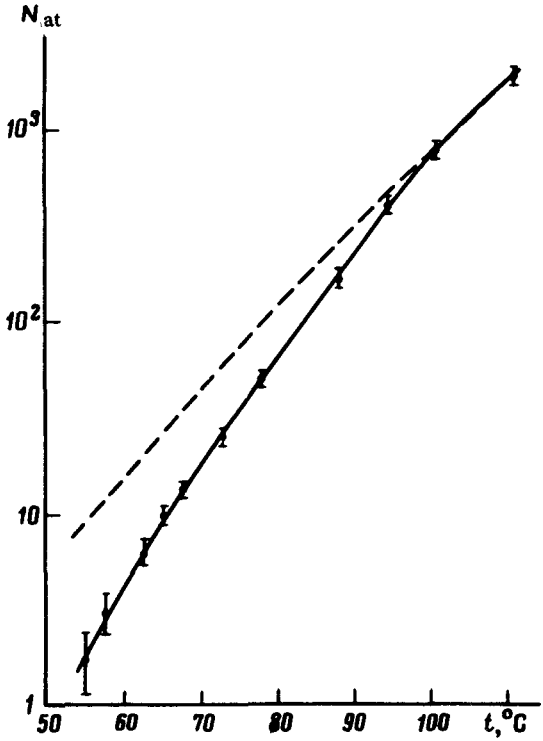

FIG. 3. Dependence of the ion yield on the temperature of the sodium oven at the maximum laser-pulse energy density and at $E=17$ kV/cm.

in the number of sodium atoms in the excited volume, which become of the same order with the average number of atoms in the value at low beam density.

4. Although the method proposed for the registration of the atoms is destructive, i.e., the registered atom is ionized and is knocked out of further interaction with the laser field, it is more universal than the method of laser fluorescent detection of atoms,[10] since Rydberg states exist for all atoms. For multiple reradiation of photons takes place in the fluorescent method, it is necessary to have a transition close to a two-level system, and this narrows down greatly the class of detected atoms.

The method of ionization of Rydberg atoms by an electric field, as shown in the present article, relaxes the requirements on the energetics of the ionizing laser radiation by 3–5 orders of magnitude, making it possible to carry out, even with the presently existing types of tunable lasers, effective detection of practically each of the atoms passing through the laser beam, for elements of the greater part of the periodic table.

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