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# Ionization detection of single atoms by laser radiation using Rydberg states

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Single atoms of sodium and ytterbium were detected by the electric-field ionization of atoms from high (Rydberg) states. Sodium and ytterbium atoms in a beam were stepwise excited to Rydberg states by radiation from tunable dye lasers and ionized by electric field pulses. The conditions for maximum ion yield were investigated and implemented in the excitation and ionization processes. The statistics of appearance of atoms in the detection zone was studied for low-density beams under ion counting conditions.

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## 1. INTRODUCTION. FORMULATION OF THE PROBLEM

Much attention has been paid recently to methods for detecting very small amounts of matter. The smallest amount of matter detectable in the atomic approach is a single atom because it carries full spectral information on matter. Consequently, the sensitivity of an ideal recording system should be capable of detecting a single atom. Among all the detection methods, the most promising is the laser excitation of resonance fluorescence and selective stepwise ionization of atoms by laser radiation.

In the resonance fluorescence method,<sup>1</sup> an atom emits photons repeatedly, returning to the initial state after each emission and it can be subjected once again to laser radiation. However, repeated excitation and relaxation of an atom back to the ground state require the existence of a transition close to that found in a two-level system. This seriously restricts the class of atoms that can be detected. The stepwise ionization method<sup>2,3</sup> is more universal than the fluorescence detection technique but it is destructive because the recorded atom is ionized and no longer interacts with the laser field. Attention has been drawn<sup>4-6</sup> to the possibility of using the stepwise ionization method in detecting atoms since the quantum efficiency of photoionization is close to unity if the energy density of laser pulses is high enough.<sup>7</sup> Modern methods for recording charged particles can detect ions and electrons with near-unity efficiency. If the transitions involved in the excitation stages are completely saturated, it is possible to use about half the atoms in the ground state. Consequently, under these conditions, one can detect every second atom. The first successful experimental detection of single atoms by the stepwise photoionization method was

reported in Ref. 8. Cesium atoms in a buffer gas were excited to the  $7^2P_{3/2}$  state by dye laser pulses and the same pulses were used for the subsequent ionization. The resultant electron-ion pairs were detected by a proportional counter. The system thus provided a stable means for detecting single cesium atoms.

However, the application of this method to atoms moving at thermal velocities in vacuum, which are of the greatest interest, presents the difficulty of very stringent requirements in respect of the energy of ionizing laser radiation. This is due to the relatively small photoionization cross section of the atoms ( $\sigma_{\text{ion}} \approx 10^{-17} - 10^{-18} \text{ cm}^2$ ). When the thermal velocity is  $\bar{v} = 5 \times 10^4 \text{ cm/sec}$  and the diameter of a laser beam is  $d = 1 \text{ cm}$ , every atom moving at right-angles to a laser beam can be ionized if the laser pulses are repeated at a frequency of 50 kHz. Under these conditions, the average laser radiation power required for efficient ionization amounts to several kilowatt.

The energy of laser pulses needed for the ionization of an atom can be reduced by several orders of magnitude if an atom is ionized from a high state by an electric field. This ionization method was proposed, implemented, and investigated in detail by the present authors and their colleagues.<sup>9-11</sup> In this method, the nonresonance process of photoionization from an intermediate state is replaced by resonance excitation of an atom to a high (Rydberg) state and subsequent ionization by an electric field pulse, which ensures near 100% ion yield.

An atom can be excited to a high state in two or three stages by radiation from several pulsed dye lasers synchronized with one another. The selection of the excitation scheme depends on the actual atom. For alkali metal atoms, it is convenient to use the two-stage excitation scheme. In the case of heavy elements with

complex atomic spectra and ionization potentials exceeding 6 eV, it is preferable to use a three-stage excitation scheme to higher states. Since the processes of stepwise excitation of an atom to Rydberg states are of resonance type, saturation requires relatively low energy densities in laser pulses, which can be attained using the available dye lasers.

We investigated the process of selective excitation of atoms to Rydberg states by the radiation of pulses dye lasers and electric-field ionization of highly excited atoms as a means for detecting single atoms. The investigation was first carried out on a fairly simple atom with one electron in excess of the filled shells (sodium) and the method was then applied to a lanthanide (ytterbium).

## 2. EXPERIMENTAL METHOD AND DESCRIPTION OF APPARATUS

The binding energy of an outer electron of a highly excited atom is a few hundredths of an electron volt and, therefore, an atomic beam is used to avoid collisional ionization of Rydberg atoms. The atoms in a beam are excited to Rydberg states in two or three stages by the radiation of pulsed dye lasers with continuously tunable frequencies. Laser beams intersect an atomic beam between two electrodes to which an electric field pulse is applied. The resultant ions are extracted through a slit in one of the electrodes and are detected with a secondary-electron multiplier. Figure 1 shows schematically the apparatus used in the stepwise excitation of atoms to high states and subsequent electric-field ionization. A transverse-discharge nitrogen laser 1 (output power per pulse 350 kW, pulse duration at midamplitude 10 nsec, repetition frequency 12 Hz), excited simultaneously two or three dye lasers. The latter consisted of the following elements: a mirror with a reflection coefficient 4–20%, a cell with a static dye solution, a telescopic expander with 25-fold magnification, and a diffraction grating with 1200 lines/mm in the first order. The width of the dye laser emission spectra was  $\Delta\nu_{\text{las}} \approx 1 \text{ cm}^{-1}$  and the duration of the dye laser pulses was  $\tau = 7 \text{ nsec}$ .

The dye laser beams were directed to a vacuum chamber where they intersected an atomic beam between two electrodes. The residual pressure in the chamber

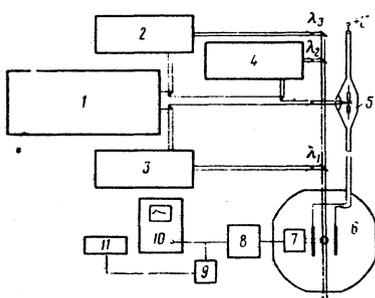


FIG. 1. Schematic diagram of the apparatus: 1) nitrogen laser; 2)–4) dye lasers; 5) generator of electric field pulses; 6) vacuum chamber; 7) secondary-electron multiplier; 8) preamplifier; 9) pulse discriminator-shaper; 10) oscilloscope; 11) frequency meter.

did not exceed  $10^{-6}$  Torr and the concentration of atoms in the excitation zone was varied by a heater and could reach  $10^8 \text{ cm}^{-3}$ .

After excitation of the atoms to a high state by the dye laser pulses, an electric field pulse of rectangular shape was applied to the electrodes. Such pulses were generated by a matched spark gap with a cable line triggered by the nitrogen laser radiation. The system made it possible to generate, across a matched load, a single rectangular voltage pulse of duration 10 nsec and up to 20 kV in amplitude, which was synchronized to within 5 nsec with the nitrogen laser pulse. The duration of the cable transmission line was selected so that the voltage pulse appeared across the electrodes 20 nsec after the laser pulses.

The ions formed as a result of ionization of the Rydberg atoms by the electric field pulse (Fig. 2) acquired a horizontal velocity component in the same pulse. The value of this component was two orders of magnitude higher than the average velocity of an atom in a beam and, therefore, the ions moved practically normally to the atomic beam. Having reached the electrode with zero potential, the ions were extracted through a slit in this electrode by the electric field of the cathode of a secondary-electron multiplier to which a potential of  $-4 \text{ kV}$  was applied. The maximum energy acquired by an ion in the region between the electrodes did not exceed 0.3 keV and, in the multiplier field, it became 4 keV. Therefore, the change in the ion energy due to a change in the electric field intensity had practically no effect on the output characteristics of the multiplier. The excitation (or detection) zone, i.e., the zone where the laser beams intersected the atomic beam, was a cylinder, 0.3–1.0 mm in diameter and 3–5 mm long. The dimensions of the slit used to extract the ions were  $3 \times 10 \text{ mm}$ . Thus, the geometry of the electrodes, slit, and excitation zone ensured that practically all the ions were extracted from the gap between the electrodes. The signal was maximized by spatial alignment of the multiplier relative to the beam. Under these conditions, the efficiency of ion detection by the multiplier was close to unity.<sup>12,13</sup>

The multiplier signal was amplified and displayed on the screen of an oscilloscope 10 or applied to a pulse-counting system comprising a pulse discriminator-shaper 9 and a frequency meter 11.

Figure 3 shows the energy level schemes of sodium (a) and ytterbium (b) atoms together with the transitions used in the excitation process. The sodium atoms were excited from the ground  $3^2S_{1/2}$  state to the resonance

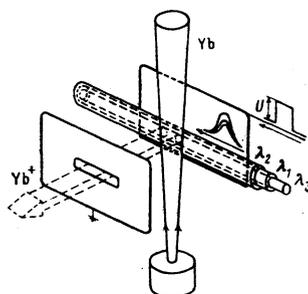


FIG. 2. Relative positions of laser beams and atomic beam of ytterbium.

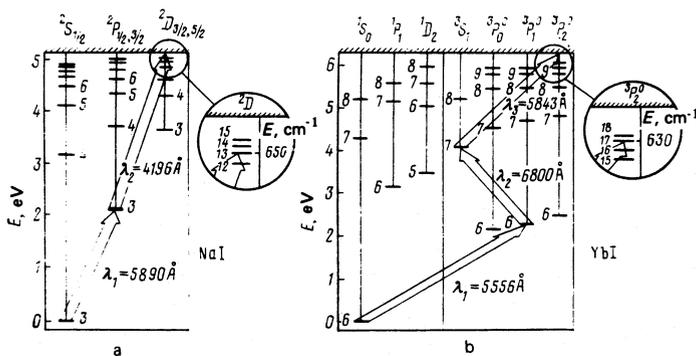


FIG. 3. Energy level schemes of sodium (a) and ytterbium (b) atoms, and transitions employed.

$3^2P_{3/2}$  state by the first-stage laser emitting at the wavelength of 5890 Å. The wavelength of the second laser was tuned within the range 4245–4160 Å, which made it possible to excite the *S* and *D* Rydberg states of the sodium atom (with the principal quantum number  $n = 12-16$ ) from the state  $3^2P_{3/2}$ . A three-stage excitation scheme was used in the case of ytterbium. The first-stage laser emitted at  $\lambda_1 = 5556.5$  Å and transferred ytterbium from the ground state  $6^1S_0$  state to the  $6^3P_1^0$  state. The second laser, emitting at  $\lambda_2 = 6799.6$  Å, excited the atoms further to the  $7^3S_1$  state. Tuning of the third laser in the range 5950–5770 Å made it possible to excite the ytterbium atoms to *P* states with  $n = 14-20$ .

### 3. MAXIMIZATION OF THE ION YIELD

The dependence of the ion yield on the electric field intensity was investigated for high *P* ytterbium states with the principal quantum number  $N = 16-20$ . The results yielded characteristic threshold curves, similar to the dependences obtained by us for sodium and rubidium.<sup>11,14</sup> Figure 4 shows the dependence of the critical electric field (deduced from the threshold curve at the ion signal level at half the saturation value) on the effective principal quantum number  $n^*$ . The critical field of ytterbium was, as in the case of the elements investigated earlier, proportional to  $(n^*)^4$ . This confirmed the universality of the proposed ionization method.

An important factor was the choice of the principal

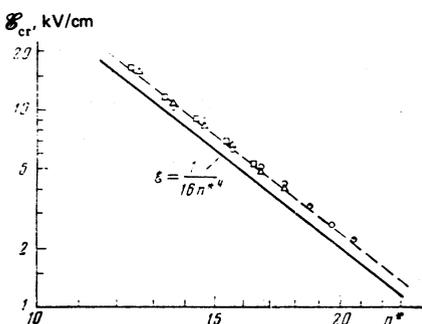


FIG. 4. Dependence of the critical electric field  $\mathcal{E}_{cr}$  on the effective principal quantum number  $n^*$  of the excited level:  $\Delta$ ) results of Ref. 11 for sodium (*S* states);  $\circ$ ) results of Ref. 14 for rubidium (*D* states);  $\square$ ) our results for ytterbium (*P* states).

quantum number  $n$  of the Rydberg state. Efficient excitation of atoms to higher states required saturation of the selected transition. The excitation cross section of an atom in the next stage fell on increase in the principal quantum number  $n$  of the level. On the other hand, the critical field rose steeply on reduction in  $n$ . The optimal case corresponded to the maximum possible excitation cross section of a high state and an electric field (required for effective ionization) of intensity easily attainable under laboratory conditions. These conditions were satisfied by the  $13^2D_{5/2}$  state of sodium ( $\mathcal{E}_{cr} = 14.2$  kV/cm) and the  $17^3P_2^0$  state of ytterbium ( $\mathcal{E}_{cr} = 11.5$  kV/cm).

Figures 5a–5c show the dependences of the ytterbium ion yield on the energy density of the laser pulses in the first, second, and third excitation stages (similar dependences were obtained for sodium for the two excitation stages employed). These dependences were then used to find the energy density of the laser pulses needed to saturate the selected transitions. These were  $E_1 = 8 \times 10^{-5}$ ,  $E_2 = 10^{-6}$ , and  $E_3 = 3 \times 10^{-3}$  J/cm<sup>2</sup>, respectively. The energy density corresponding to the saturation of a transition can be estimated from

$$E_{sat} = \frac{h\nu}{2\sigma}, \quad \sigma = \frac{g_b \lambda^2}{g_a 2\pi} \frac{A_{ba}}{\Delta\omega},$$

where  $\sigma$  is the cross section of the  $a \rightarrow b$  radiative transition;  $h\nu$  and  $\lambda$  are the quantum energy and wavelength of the transition;  $g_b$  and  $g_a$  are the degeneracy factors of the upper and lower levels;  $A_{ba}$  is the Einstein coeffi-

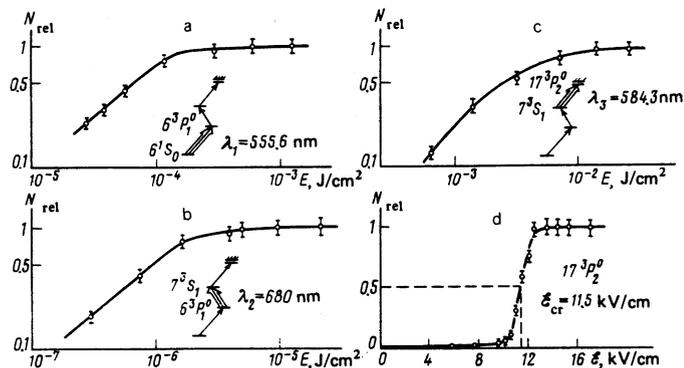


FIG. 5. a)–c) Dependences of the ytterbium ion yield on the energy density of laser pulses in the first, second, and third excitation stages, respectively, for an electric field of  $\mathcal{E} = 14$  kV/cm. d) Dependence of the ion yield on the electric field intensity for the  $17^3P_2^0$  state.

cient;  $\Delta\omega$  is the width of the spectral line of the transition. The Einstein coefficients of the  $6^3P_1 \rightarrow 6^1S_0$  and  $7^3S_1 \rightarrow 6^3P_1$  transitions are well known:  $A_{6P-6S} = 1.2 \times 10^6 \text{ sec}^{-1}$ ,  $A_{7S-6P} = 10^8 \text{ sec}^{-1}$ . The width of the line  $\Delta\nu_{\text{abs}}$  representing absorption in the atomic base used in our experiments is  $0.01 \text{ cm}^{-1}$ , and the corresponding transition cross sections are  $\sigma_{6S-6P} = 1.5 \times 10^{-12} \text{ cm}^2$  and  $\sigma_{6P-7S} = 6 \times 10^{-11} \text{ cm}^2$ . The energy densities of the laser pulses needed to saturate the transitions are

$$E_i = E_{i, \text{sat}} \frac{\Delta\nu_{\text{las}}}{\Delta\nu_{\text{abs}}}$$

and, for the laser radiation with  $\Delta\nu_{\text{las}} = 1 \text{ cm}^{-1}$ , they are  $E_1 = 2 \times 10^{-5}$  and  $E_2 = 5 \times 10^{-7} \text{ J/cm}^2$ , respectively. The discrepancy between the experimental and theoretical results is clearly due to inaccurate determination of the transverse dimensions of the laser beams. We used the curves in Figs. 5a-5c to find the working energy densities of laser pulses for all the excitation stages and this was done so as to ensure saturation of the absorption in all three transitions at the same time. The populations of the excited levels then obeyed the following relationships:

$$\frac{n_1}{n_0} = \frac{g_1}{g_0}, \quad \frac{n_2}{n_1} = \frac{g_2}{g_1}, \quad \frac{n_3}{n_2} = \frac{g_3}{g_2} \quad \text{and} \quad n_0 + n_1 + n_2 + n_3 = N,$$

where  $n_i$  and  $g_i$  ( $i = 0, 1, 2, 3$ ) are the population density and degeneracy factor of the  $i$ th level;  $N$  is the density of atoms in the excitation zone. These relationships yield the following expression for  $n_3$ :

$$n_3 = Ng_3 / (g_0 + g_1 + g_2 + g_3).$$

Thus, about five-twelfths of all the atoms in the excitation zone were excited to the  $17^3P_2^0$  state. In the case of sodium, the proportion of highly excited atoms ( $13^2D_{5/2}$ ) state was one-half. Figure 5d shows the dependence of the ytterbium ion yield on the intensity of the electric field pulses used to excite them to the  $17^3P_2^0$  state. Saturation of the ion signal in the ionization stage appeared during the field pulse for all the atoms excited to the Rydberg state. The working electric field intensity was selected to be  $14 \text{ kV/cm}$  ( $17 \text{ kV/cm}$  for sodium). Thus, about half the atoms that were in the detection zone at the moment of arrival of the laser pulses were excited to a high state and each highly excited atom was ionized.

#### 4. TEMPERATURE DEPENDENCE OF THE ION SIGNAL AND ITS CALIBRATION

Figure 6 shows the dependence of the number of atoms in the detection zone on the temperature of the heater calculated for an ytterbium beam in the standard way on the basis of the results of Ref. 15 (dashed curve). This figure also includes the dependence of the ytterbium ion yield on the temperature in the heater (continuous curve) obtained for working values of the energy densities in the laser pulses and the intensities of the electric field pulses. The experimental and calculated curves are parallel at heater temperatures in excess of  $250^\circ\text{C}$ . This makes it possible to calibrate the experimental dependence so that it coincides with that calculated for temperature in excess of  $250^\circ\text{C}$ . The discre-

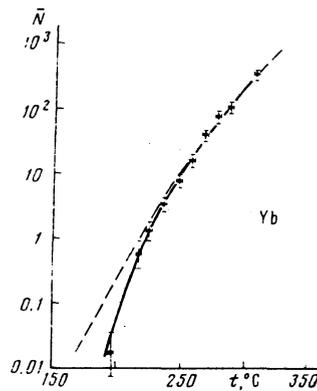


FIG. 6. Dependence of the ytterbium ion yield on the heater temperature for working values of the energy density of laser pulses and electric field intensity.

pancy between the experimental and calculated values at lower temperatures is clearly due to the fact that the atomic vapor inside the heater is not saturated because of the loss of atoms to the heater walls. The gradual fall in the ion signal amplitude on reduction in the heater temperature was observed down to about  $230^\circ\text{C}$ . Further cooling resulted in a strong instability of the signal. In the range  $220\text{--}230^\circ\text{C}$ , no ion signal was produced by the majority of the laser pulses but the amplitude of the minimum ion signal did not decrease. The ytterbium ions were detected under conditions of practically complete absence of background ion or dark pulses from the secondary-electron multiplier. Therefore, the strong instability of the ion signal was due to fluctuations in the number of atoms in the excitation zone, which could be (in the case of a low-density beam) of the same order as the average number of atoms in the zone. In fact, the calculated dependence indicated that the average number of atoms in the excitation zone at  $220\text{--}230^\circ\text{C}$  was close to unity. Significant fluctuations in the ion signal began to appear at the same temperatures. This confirmed the validity of our calibration of the experimental dependence on the basis of the calculated dependence. Thus, at the moment of arrival of the laser pulses, the excitation zone could contain no atom, one atom, two atoms, and, less likely, three, four, etc. atoms. The absence of the ion signal was then due to the absence of atoms in the excitation zone and the instability of the amplitude was due to fluctuations in the numbers of atoms in this zone and the constant minimum signal corresponded to the detection of a single atom. This behavior of the ion signal was also observed in the detection of sodium atoms.<sup>16</sup>

#### 5. FLUCTUATIONS IN THE NUMBER OF ATOMS IN THE EXCITATION ZONE

The density of an atomic beam can be made so low that the probability of simultaneous arrival of two atoms in the excitation zone is much less than the probability of arrival of a single atom. Under these conditions, the detection system should respond in most cases to just one atom. The probability of recording  $k$  atoms in a time interval  $\Delta T$  is then given by the Poisson distribu-

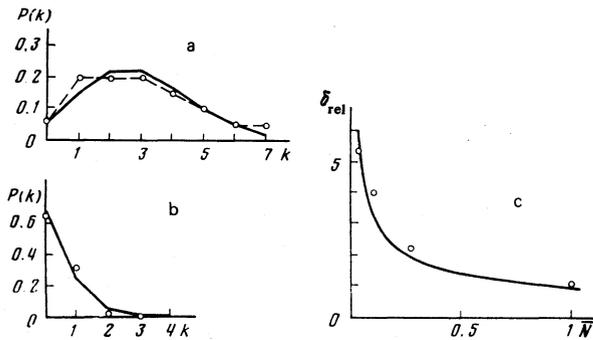


FIG. 7. Distribution of the number of ions recorded by a detection system in a time  $\Delta T$  for an average number of detected atoms and an average number of atoms in the excitation zone  $\bar{N}$ : a) sodium,  $\Delta T=20$  sec,  $\bar{k}=3$ ,  $\bar{N}=0.03$ ; b) ytterbium,  $\Delta T=20$  sec,  $\bar{k}=0.35$ ,  $\bar{N}=0.003$ ; c) dependence of the relative fluctuation in the number of ions  $\delta_{rel} = \Delta\bar{k}/\bar{k}$  on the average number of atoms  $\bar{N}$ .

tion

$$P(k) = \frac{\bar{k}^k}{k!} e^{-\bar{k}},$$

where  $\bar{k}$  is the average number of ions recorded in a time interval  $\Delta T$ .

Figures 7a and 7b give the probabilities of recording  $k$  ions in a time  $T$ , obtained experimentally for beams of sodium (a) and ytterbium (b) atoms with different values of the average number of atoms  $\bar{N}$  in the excitation zone; the latter number was governed by the temperature of the heater. For comparison, this figure also includes the Poisson distributions (continuous lines) calculated for the corresponding values of  $\bar{k}$  found experimentally. Such distributions were investigated for several values of  $\bar{N}$  for each of the atoms. Figure 7c shows the dependence of the relative fluctuation in the number of ions  $\delta_{rel} = \Delta\bar{k}/\bar{k}$  on the average number of atoms  $\bar{N}$ . The continuous curve is the same dependence as that for the Poisson distribution  $\delta_p(\bar{k}) = 1/\bar{k}^{1/2} \propto 1/\bar{N}^{1/2}$ .

It is clear from Fig. 7 that the experimental distributions are close to the Poisson curves and, consequently, the detection system does react accurately to the appearance of a single atom in the detection zone. The average number of ytterbium atoms  $\bar{N}$  corresponding to the minimum heater temperature (Fig. 6) is calculated from the experimental value of  $\bar{k}$  obtained in the ion-counting regime. This value is in good agreement with the rest of the experimental curve, which confirms once again that the system can detect single atoms.

## 6. CONCLUSIONS. APPLICATIONS

The proposed method of ionization of highly excited atoms by an electric field is thus universal and—as shown above—reduces by several orders of magnitude the requirements in respect to the energy of ionizing laser radiation. This means that even the existing types of tunable dye lasers with a low pulse repetition frequency can be used efficiently to detect atoms intersecting a laser beam under laboratory conditions. The use of a pulsed copper vapor laser (pulse repetition fre-

quency in excess of 20 kHz) in pumping dye lasers makes it possible to ionize practically every second atom intersecting the detection zone. The use of dye lasers with an emission line width of the order of the width of the absorption line of an atomic beam allows a further reduction in the laser pulse energy by one or two orders of magnitude. The energy density required for the most difficult last stage of excitation is then  $3 \times 10^{-5}$  J/cm<sup>2</sup> and the average density of the laser power emitted from a dye laser with a pulse repetition frequency of 20 kHz does not exceed 1 W/cm<sup>2</sup>, which is an easily attainable value. Apparatus based on this principle solves the problem of a laser detector of atoms of most elements in the Mendeleev periodic system.

Undoubtedly, the laser photoionization method of detecting single atoms will find extensive applications in various branches of physics and technology, such as detection of impurities in ultrapure materials, accurate measurement of the relative abundances of isotopes, etc. We should mention particularly the possibility of using this method in nuclear physics. A laser photoionization detector can be used to measure nuclear deformation in atoms available in very limited numbers. The existing laser technology can be used to determine the shifts of the atomic absorption lines of less than  $10^{-4}$  cm<sup>-1</sup>. Therefore, mean-square relative changes in the nuclear radius of the order of  $\delta\langle r^2 \rangle/R^2 \approx 10^{-5}$  or smaller can be determined. The laser photoionization method can be used to carry out these measurements when only  $10^3$ – $10^5$  atoms are available. Stepwise excitation of an atom can ensure selectivity at each stage if the transitions are selected correctly. If the atomic line shift is a few reciprocal centimeters, the selectivity in the last excitation stage is  $10^5$ – $10^{10}$ . Multiplying these two quantities, we find that the excitation selectivity of the resultant multistage process is  $10^{-16}$ – $10^{-20}$ .

Investigations of the shift of the atomic absorption line with this relative sensitivity and on such small amounts of matter should make it possible to determine the radii of the isotopes and isomers with an excess or deficiency of neutrons, which appear on approach to the instability limits. It would be interesting to use this method in searching for "exotic" nuclei such as superdense species<sup>17</sup> or in determining the existence of nuclear spontaneously fissioning shape isomers.

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