V. S. Letokhov. *Detection of single atoms and nuclei by methods of laser spectroscopy.* Considerable attention is now being given to development of methods for laser detection of ultrasmall or "trace" amounts of substances (see the review in Ref. 1). In principle, the limit of detection is the single atom, since it still carries complete spectral information on its structure. It is for this reason that one of the main objectives in laser spectroscopy is the development of methods for detection of single atoms. The most promising methods are laser excitation of resonant fluorescence, in which the largest number of photons is scattered by a single atom,[2] and selective stepwise ionization by laser light, which converts practically all of the atoms to ions.[3] Both methods have recently been domonstrated successfully at the USSR Academy of Sciences Institute of Spectroscopy.

A peculiarity of the resonant-fluorescence method is that a given atom can interact many times with the laser radiation, reradiating photons of the exciting frequency in all directions. The populations of the ground and excited levels become equal if the intensity of the laser radiation is higher than the saturation intensity of the resonant transition. In this case, the atom reradiates $N_{max} = T/2\tau_{spont}$ photons during the time T in which it crosses the light beam; here τ_{spont} is the spontaneous time of decay of the excited state to the ground state.

In the experiment of Ref. 2, for example, Na atoms crossed the light beam of a continuous dye laser whose frequency was tuned to the D_2 resonance line. Each atom reradiated about 200 photons into 4π sr on a 4-mm path. This fluorescence signal was registered by two photomultipliers (PM), and a two-channel registration scheme operating in the coincidence mode was used to extract the fluorescence signal from each atom that crossed the light beam. The lowest detectable flux of Na atoms was 10 atoms/sec, which corresponds to an average density of 10^{-4} atom in the region of registration. Unfortunately, the fluorescent method for detection of single atoms cannot be used for most complex atoms with metastable states near the ground state, since the process cycle is easily interrupted after transition of the atom from the excited to the metastable state. In this case, the photoionization method is more efficient and universal.

This approach to the detection of single atoms is based on selective stepwise photoionization, a method proposed by the author of the paper both for separation of isotopes and for detection of atoms back in 1969 (see the review in Ref. 4). If detection occurs in a vacuum or a gas at low pressure (the most interesting case from the standpoint of maximum spectral resolution), ionization of each atom that has entered the laser beam requires sending laser pulses at a repetition frequency of about 50 kHz (at a thermal velocity of 5×10^4 cm/sec and an interaction path of 1 cm). At the 1 W average power available from laboratory tunable lasers, effective ionization of most atoms can be brought about only if the atom is ionized via overlying (Rydberg) states. In this method of selective ionization, which was also proposed and investigated at the USSR Academy of Sciences Institute of Spectroscopy,[5] the process of nonresonant photoionization of the atom on the transition from the intermediate state to the continuum is replaced by resonant excitation of the atom from the same state to a higher state near the ionization limit, with subsequent ionization by an electric-field pulse. In this case the efficiency of ionization approaches unity. Since excitation is resonant at all subsequent steps, saturation of all transitions requires a comparatively small laser-pulse energy density (10^{-4} to 10^{-6} J/cm^2), which is quite within reach of most existing pulsed dye lasers.

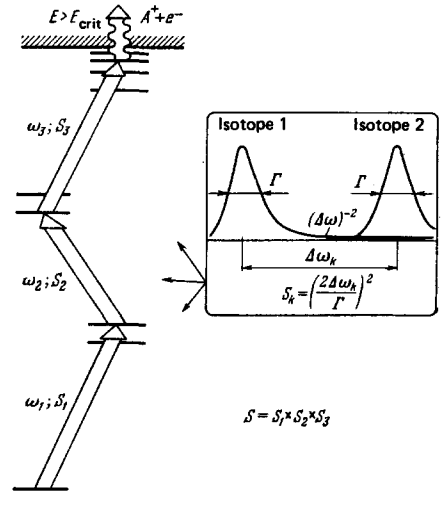

FIG. 1. Diagram of highly selective three-step excitation of a Rydberg state of an atom near the ionization limit with subsequent ionization by an electric-field pulse; used to overcome ionization-selectivity limit due to wing overlapping of closely spaced absorption lines.

This was demonstrated successfully in experiments to detect single atoms of Na^3 (two-stage laser excitation + ionization by electric field) and Yb^6 (three-stage laser excitation + ionization by electric field).

We draw attention to the fact that multistage laser excitation using the isotopic (or isomeric, etc). shift on all subsequent transitions can also be used to obtain exceptionally high selectivity in the detection of atoms (Fig. 1). The selectivity of excitation at one step is usually limited by the inevitable contribution of excitation on the Lorenz wing of an adjacent line. At best, the selectivity limit $S_1 = (2\Delta\omega_1/\Gamma)^2 \approx 10^5 - 10^6$ at a natural linewidth Γ and a distance $\Delta\omega_1$ between lines. In three-step selective excitation, however, it can be increased to $S = S_1 \times S_2 \times S_3 \approx 10^{15} - 10^{18}$. This is of fundamental importance for detection of rare isotopes, form isomers of nuclei, ultradense nuclei, etc.

The high sensitivity and resolution of the laser-spectroscopy techniques that have been developed make it possible to investigate characteristics of nuclei that are available in very small quantities. The basis for this possibility is that many characteristics of the nuclei are quite explicitly manifest in subtle details of the atom's optical spectrum: numbers of protons and neutrons, spin and quadrupole moment of the nucleus and the form associated with them, average radius and excitation of nucleus, and even its velocity of motion and and orientation. This is the key to application of laser-spectroscopy methods in nuclear-physics experiments in which operations with nuclei having the desired characteristics are accomplished by the action of coherent light on the electron cloud surrounding the nucleus.

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