

Laser light, atoms, and nuclei

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A brief review is given of recent work on the application of laser radiation in nuclear physics, including the laser spectroscopy of unstable nuclei, laser detection of very rare nuclei, laser orientation and separation of nuclei, mixed nuclear-optical  $\gamma$ -transitions, and other topics.

INTRODUCTION

In-depth studies of the structure of matter clearly demonstrate that there are three principal subdivisions in physics:<sup>1</sup> (1) atomic physics, whose concern is the structure of the electron shells of atoms and the structure of molecules, (2) nuclear physics, which deals with the structure of the nucleus as a system of nucleons, and (3) high-energy physics, i.e., the study of the structure of nucleons and of other elementary particles. There are remarkably profound analogies<sup>2</sup> between these apparently disparate fields of study, which have become particularly obvious in recent years as great advances were being made in elementary-particle physics (electroweak interaction and quantum chromodynamics).

The three ranges of transition energies corresponding to the atom, the nucleus, and the nucleon have associated with them three types of spectroscopy<sup>1,2</sup> that have gradually evolved in the last hundred years: (1) optical spectroscopy of atoms and molecules, (2) nuclear spectroscopy of transitions in the nucleus, and, finally, (3) the spectroscopy of elementary particles and of the quark-quark interaction (charmonium, etc.). These three spectroscopies occupy energy ranges (interaction energies of electrons in the atom, nucleons in the nucleus, and quarks in the nucleon) that differ from each other by 4–6 orders of magnitude (Fig. 1). The three branches of physics and the three spectroscopies can therefore be looked upon, in the first approximation, as completely independent of one another. There is, however, a number of effects relating, say, the electron shells of the atom with nuclear characteristics, and even with interactions in the interior of the nucleon. They are the basis for the connection between atomic physics and optical spectroscopy, on the one hand, and nuclear physics and even elementary-particle physics, on the other.

1. CONNECTIONS BETWEEN ATOMIC AND NUCLEAR PHYSICS

Consider a nucleus surrounded by the electron shell of an atom or ion, which can also be part of a molecule or molecular ion. All the effects that we shall examine are based on the fact that laser light can interact with the electron shells of atoms (in some cases, with molecular vibrations as well). Since these shells are coupled to the nucleus, the effects can be used to obtain information on the characteristics of the nucleus and on the fundamental interactions within it, to discover new nuclei, to influence processes in which they participate, and so on.

The first and most obvious interaction between the nucleus and the electron shell of the atom is the *Coulomb* interaction that ensures their permanent mutual proximity. This interaction is sensitive to the nuclear charge  $Z$ , and this manifests itself in the optical spectrum of the atom. It provides the basis for the detection of nuclei of a particular element by any of the methods employed in optical spectroscopy. This basic possibility had not been used in nuclear physics but, with the advent of the ultrasensitive laser methods for the detection of single atoms,<sup>3,4</sup> it has become very promising for nuclear physics.

The interaction energy between an optical electron and a nucleus depends to a small extent on the magnetic moment  $\mu$  and the quadrupole moment  $Q$  of the nucleus (*hyperfine interaction*), and this manifests itself in the hyperfine structure of optical spectral lines.<sup>5</sup> This has long been used in systematic determinations of the moments of stable nuclei,

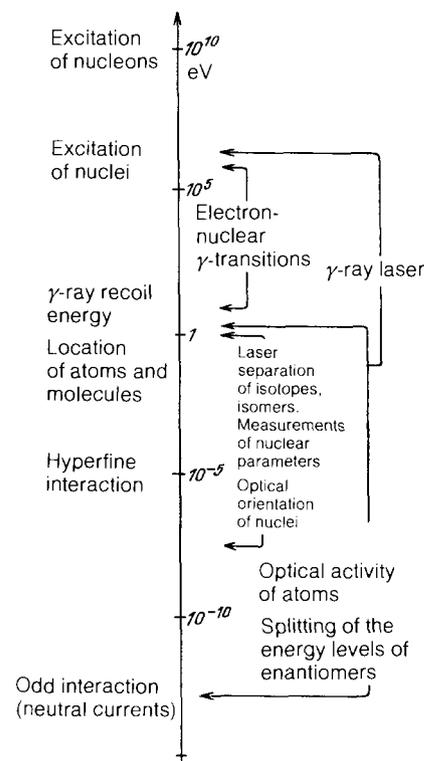


FIG. 1. Energy scale showing atomic, nuclear, and nucleonic excitation energy ranges together with the interaction energies at the interfaces between atomic physics, nuclear physics, and elementary-particle physics. The various effects based on these interactions are shown on the right.

using atomic spectra.<sup>6</sup> The hyperfine interaction can also be used to orient nuclei by exciting an optical electron with circularly polarized light (optical pumping<sup>7</sup>), and is the basis of techniques employed to polarize nuclei in vapors and in atomic beams for nuclear-physics experiments.

A change in the number  $N$  of neutrons in the nucleus (isotopy) produces a change in atomic and, especially, molecular spectra, since it gives rise to a small change in the electric field of the nucleus that acts on the electrons, and to a change in the nuclear mass. The isotopic field effect provides information on variations in the charge radius  $\delta\langle r \rangle$  of the nucleus as the nucleon number  $N$  is varied. Moreover, a change in  $N$  and the excitation of a nucleus produce a change in its moments and, hence, have a definite effect on hyperfine structure.

Hyperfine interactions and isotopic effects constitute a bridge between energy ranges separated by 4–6 orders of magnitude ( $10^{-4}$ – $10^{-5}$  and 1–10 eV, respectively). A weaker connection is also possible. For example, the excitation of a nucleus produces a change in its mass by  $0.1 m_e - m_e$ . This should result in an isomeric mass shift of  $10^{-10}$  eV, which is negligible.

When used in combination with methods capable of ultrahigh sensitivity, high-resolution laser spectroscopy (including laser spectroscopy without Doppler broadening<sup>8</sup>) is of great value in studies of the characteristics of nuclei, especially short-lived unstable nuclei that are available in only very small amounts. From the standpoint of nuclear physics, the success of laser optical spectroscopy in the study, detection, etc. of nuclei is based on two properties: (1) the large cross section for the resonance excitation of optical transitions in the electron shell ( $\sigma_{\text{opt}} \approx \lambda^2/2\pi \approx 10^{-10}$  cm<sup>2</sup> =  $10^{-14}$  barn) and (2) the high intensity of even relatively modest laser beams (1 W/cm<sup>2</sup> corresponds to an intensity  $I \approx 10^{19}$  photons/cm<sup>2</sup>s). This ensures the high rate of resonance excitation  $W_{\text{exc}} = \sigma_{\text{opt}} I \approx 10^9$  s<sup>-1</sup> of atoms in a beam for given nuclear charge  $Z$  and given neutron number  $N$ . Moreover, the scope of laser methods is actually much more extensive: it is possible to achieve not only isotopically, but also isomerically selective excitation of atoms, which means that isotopic and isomeric nuclei can be detected and separated by laser radiation.<sup>9</sup>

Fundamental interactions between elementary particles within the nucleus influence the optical spectra of atoms and molecules, but the effect is exceedingly small. In particular, parity violation in *weak interactions* that conserve nuclear charge give rise to certain effects in atomic and molecular spectra<sup>10</sup> (optical activity in isotropic media<sup>11</sup> and level splitting in left-handed and right-handed molecules<sup>12</sup>). These effects are very small but, again, they provide a bridge between very distant energy ranges (between 1 eV and  $10^{-10}$ – $10^{-15}$  eV; see Fig. 1).

The nucleus and the electron shell of an atom move together as one whole. This gives rise to *kinematic effects* that connect optical spectra, on the one hand, and nuclear characteristics, on the other. Thus, first, the nucleus and the electron shell exhibit the same relative Doppler frequency shift. Laser radiation can be used to excite, or even ionize, atoms of a particular velocity. This can be exploited in velocity measurements and in monochromatization (using an optical transition in the electron shell). Second, the recoil effect accompanying the absorption or emission of a  $\gamma$ -ray by

the nucleus produces a change not only in the translational energy of the nucleus, but also in the internal energy of its ambient electrons, and it may even modify the vibrational energy of a molecule containing this type of nucleus. This is responsible for electronic and vibrational satellites of  $\gamma$ -ray lines in the emission and absorption spectra of the nucleus.<sup>13</sup> In other words, there should be nuclear-electronic and nuclear-vibrational  $\gamma$ -transitions in nuclei, resembling the electronic-vibrational transitions in molecules. It can be used together with laser excitation to influence the structure and even the shape of a line resulting from a  $\gamma$ -transition. The technique provides a bridge between atomic and molecular excitation energies (0.1–10 eV), on the one hand, and the nuclear excitation energies ( $10^4$ – $10^6$  eV), on the other (see Fig. 1). Finally, a strong laser field may well affect the rate of processes occurring in the nuclear interior, including  $\beta$ -decay<sup>14</sup> which exhibits the greatest sensitivity to the electric field. Although these effects lie outside the range of present experimental techniques, we have, at least in principle, the prospect of a connection between high-intensity and high-energy physics.

Below, we briefly discuss the basic physical ideas, the results, and the potentialities of methods of exploring nuclei with laser light. More detailed accounts and citations of original papers can be found in published reviews<sup>15–17</sup> and proceedings of international conferences.<sup>18–19</sup>

## 2. LASER SPECTROSCOPY OF UNSTABLE NUCLEI

The techniques of atomic spectroscopy can be used to investigate nuclear parameters such as spin, electric and magnetic dipole moments, and variations in the root mean square charge radius along an isotopic chain. These nuclear parameters influence the hyperfine structure and isotopic shifts in optical transitions.

The energy of a hyperfine-structure component with total angular momentum  $F = I + J$  that is due to the interaction between the electron shell and the nucleus is given by<sup>5,6</sup>

$$W_F = \frac{1}{2} KA + \frac{(3/4)K(K+1) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)} B, \quad (1)$$

where  $K = F(F+1) - I(I+1) - J(J+1)$ . This energy depends on the nuclear spin  $I$ , the magnetic dipole interaction constant

$$A = \mu_I H_e(0) (IJ)^{-1} \quad (2)$$

and the electric quadrupole interaction constant

$$B = eQ_s \frac{\partial^2 V(0)}{\partial z^2} = eQ_s \varphi_{JJ}(0). \quad (3)$$

The nuclear moments of  $\mu_I$  and  $Q_s$  can be obtained by measuring the hyperfine structure of spectral lines (which yield the constants  $A$  and  $B$ ) and using empirical or theoretical values of the hyperfine magnetic field  $H_e(0)$  and the electric field gradient  $\varphi_{JJ}(0)$  at the nucleus.

The isotopic shift  $\delta\nu^{AA'}$  of the center of gravity of a hyperfine structure multiplet due to an optical transition is related to the change in the mean square charge radius  $\Delta\langle r^2 \rangle^{AA'}$  between isotopes  $A$  and  $A'$  by

$$\delta\nu^{AA'} = f \Delta\langle r^2 \rangle^{AA'} + M \frac{A' - A}{A'A}, \quad (4)$$

where the electron factor  $f$  in the isotopic field shift is pro-

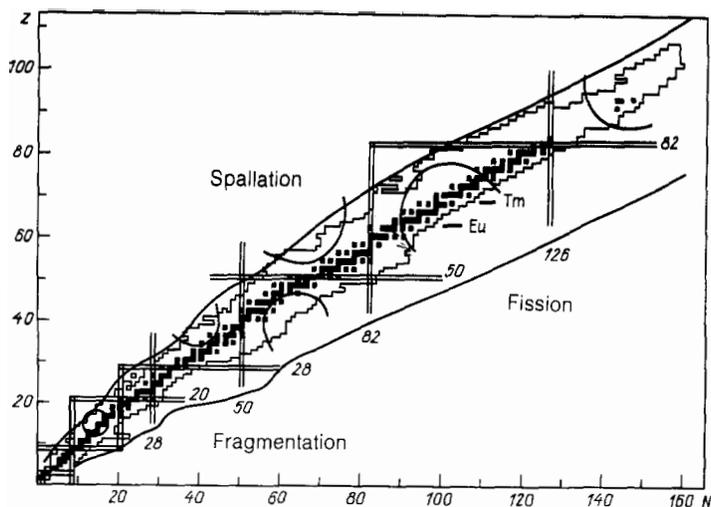


FIG. 2. Distribution of nuclides ( $Z$  is the proton number and  $N$  the neutron number of the nucleus), showing stable nuclei (black), radioactive nuclei, and magic numbers (double lines). The diagram also shows the basic nuclear reactions in which radioactive isotopes are produced.

portional to the change  $\Delta|\Psi(0)|^2$  in the electron density at the nucleus between the states:

$$f = \frac{2}{3} \pi Z e^2 \Delta|\Psi(0)|^2, \quad (5)$$

where  $\Psi(0)$  is the amplitude of the electron wave function at the nucleus, which can be calculated. The second term in (4) is the isotopic mass shift due to the change in the nuclear recoil energy. For simple  $s \rightarrow p$  or  $s^2 \rightarrow sp$  electron transitions in heavy atoms, the second term is small in comparison with the first.

The techniques of optical spectroscopy have long been used with considerable success in determinations of  $I$ ,  $\mu$ ,  $Q$  and  $\Delta\langle r^2 \rangle$  of stable isotopes available to experimenters in sufficient amounts.<sup>6</sup> The last decade has opened up the possibility, at least in principle, of systematic measurements on unstable (including short-lived) isotopes lying in the wings of the  $\beta$ -stability region (Fig. 2). Such isotopes are produced in nuclear reactions that occur when targets are exposed to accelerated protons, ions, or neutrons (spallation, fragmentation, and fission reactions; see Fig. 2). The new nuclei produced in the target can be evaporated out of the latter, separated in accordance with their masses in a magnetic mass separator, neutralized by, say, implantation in a foil, and finally released in the form of a beam of neutral atoms when the foil is heated. This is illustrated schematically in Fig. 3. Radioactive isotopes can be used in two ways: (a) off-line,

i.e., by first storing the isotopes and then releasing them for measurement, which can be used with relatively long-lived isotopes (half-lives of hours, days, or weeks), and (b) on-line, suitable for short-lived isotopes with half-lives of fractions of a second, but limited by the time of thermal diffusion of the resulting nuclei out of the target. Several mass separators have now been developed for studies of short-lived isotopes by high-sensitivity and high-resolution laser spectroscopy.<sup>20,21</sup>

The first apparatus incorporating a mass separator for the production and systematic investigation of the properties of short-lived isotopes is the ISOLDE (Isotope Separation On-line Detection).<sup>21</sup> The system produces beams of separated radioactive nuclei in amounts of  $10^8$ – $10^5$  atoms/s, which is sufficient for hyperfine-structure and isotopic-shift measurements by high-sensitivity high-resolution laser spectroscopy. ISOLDE experiments with radioactive isotopes employ many of the high-sensitivity techniques of linear and nonlinear laser spectroscopy (see the review given in Ref. 3) and have produced numerous and very interesting results. Already early experiments<sup>22</sup> with the isotopes of mercury in a resonance cell, in which hyperfine structure and isomeric shifts were investigated using the excitation of fluorescence, showed evidence of an unexpected effect, namely, an abrupt change in the shape of the nuclei of the light isotopes of mercury. Experiments with radioactive

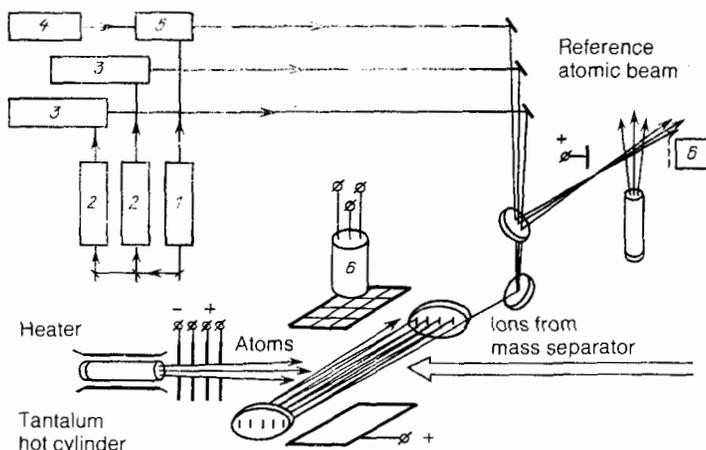


FIG. 3. Laser photoionization spectrometer of the nuclear/laser complex at the Konstantinov Leningrad Institute of Nuclear Physics and the Institute of Spectroscopy of the USSR Academy of Sciences (proton accelerator, target, and mass separator are now shown). This spectrometer is based on the method of three-step resonance ionization of atoms by radiation from three dye lasers (3–5; 4—narrow-band fixed-frequency continuously operating laser with a pulsed dye-laser amplifier (5), pumped by pulses from copper vapor lasers (1,2) with pulse repetition frequency 10 kHz, 6—secondary electron multipliers (taken from Ref. 33).

atoms in such cells (as opposed to atomic and ionic *beams*) can be used to achieve high sensitivity and high spectral resolution when combined with different Doppler-free nonlinear laser spectroscopic techniques<sup>23,24</sup> (see Ref. 8).

Magnetic deflection of atoms has been used in successful experiments with beams of radioactive isotopes of the alkali metals (in a Stern-Gerlach analyzer), oriented by resonant interaction with laser radiation<sup>25,26</sup> (see the review given in Ref. 3). High spectral resolution is achieved in experiments of this kind by crossing a collimated atomic beam with a laser beam. Systematic measurements performed by this method on ISOLDE produced interesting data on isomeric shifts, spins, hyperfine structures, and nuclear deformations along long chains of radioactive isotopes of Na, K, Rb, Cs, and Fr, produced by the mass separator at the rate of  $10^5$ – $10^{10}$  atoms/s (Refs. 27 and 28).

High spectral resolution and good sensitivity can be achieved in collinear laser spectroscopy of beams of accelerated atoms, when velocity bunching of atoms under collinear excitation removes Doppler broadening, and laser excitation of fluorescence ensures good sensitivity.<sup>29</sup> This method has been used to obtain interesting data on nuclear deformation along chains of radioactive neutron-deficient isotopes of a number of rare-earth elements (Gd, Dy, Er, Yb) and the isotopes of radium.<sup>30,31</sup>

In the USSR, the B. P. Konstantinov Institute of Nuclear Physics in Leningrad, working in collaboration with the Institute of Spectroscopy of the USSR Academy of Sciences, has developed a nuclear/laser measuring system (IRIS) that incorporates a mass separator of radioactive atoms and a laser photoionization spectrometer<sup>32,33</sup> that uses an exceptionally sensitive method of resonance step-wise ionization of atoms by laser radiation (see Ref. 4). Ions of the isotopes under investigation are produced in the tantalum target of the mass separator, bombarded by 1-GeV protons. The ions are then directed on to a heated tantalum crucible at the end of the mass separator (Fig. 3). The crucible produces (in the reverse direction) a collimated atomic

beam of the isotopes, which crosses at right angles the three coincident beams of dye-laser radiation. Higher efficiency is achieved by using two flat mirrors to reflect the laser beams several times across the atomic beam. The frequencies of the three-color laser radiation are equal to the frequencies of three successive atomic transitions. The three-step excitation takes the atom to the autoionizing state, and the resulting photoions are recorded by a secondary-electron multiplier. The optical spectrum is recorded when the number of photoions is measured as a function of the frequency of the tunable laser radiation in the first excitation step. The particular feature of this system is the use of excitation by dye lasers with high pulse repetition frequency (10 kHz), the lasers themselves being pumped by pulse-periodic copper-vapor lasers. The high laser pulse repetition frequency is necessary to "capture" the maximum number of rare atoms that rapidly cross the illuminated region. With spectral resolution of about 100 MHz (determined by the spectral linewidth of the atomic beam), the sensitivity of the photoionization method is sufficient for spectroscopic measurements on isotopes produced in the target at a rate down to 3000 nuclei/s. The flux of mass-separated ions used in these measurements is usually in the range of  $10^4$ – $10^5$  s<sup>-1</sup>.

This method has been used at the Konstantinov Institute and the Institute of Spectroscopy to investigate the isotopic shifts and hyperfine structures of the isotopes of Nd ( $A = 132, 134$ – $142$ ), Sm ( $A = 138$ – $145, 147, 149, 150, 152, 154$ ), Eu ( $A = 138$ – $151$ ), Ho ( $A = 152$ – $165$ ), and Tm ( $A = 156$ – $172$ ) (Ref. 34). These measurements have yielded the mean square charge radii of the nuclei of these isotopes, their electromagnetic moments (with the exception of the even-even isotopes and the Eu isotopes), improved values of the spins of certain nuclei, and the isotopic behavior of the charge radius  $\Delta\langle r^2 \rangle$  of rare earth elements for  $N < 82$  and  $88 < N < 94$ , i.e., on either side of the magic number  $N = 82$ . Figure 4 shows the values of  $\Delta\langle r^2 \rangle$  for the isotopes of these elements. For the isotope chains of Nd, Sm, and Eu, there is a clear shell effect, i.e., the rate of change of the

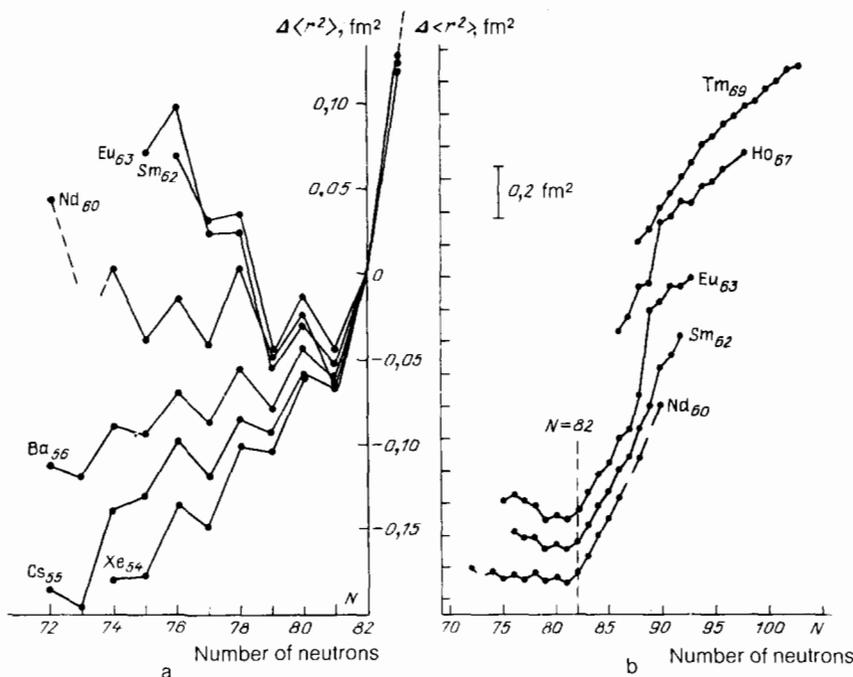


FIG. 4. Change in the mean square charge radius  $\Delta\langle r^2 \rangle$  as a function of the number  $N$  of neutrons in the nucleus: a— $N < 82$  for Sm, Eu, and Nd (data from Ref. 34) and Ba, Cs and Xe (data from Ref. 35), b— $N > 82$  for Nd, Sm, Ho, and Tm (data from Ref. 34).

radius changes at  $N = 82$ . Figure 4a shows the isotopic variation in  $\Delta\langle r^2 \rangle$  for  $N < 82$  for Eu, Nd, and Sm, and the previously investigated<sup>35</sup> isotopes of Ba, Cs, Xe (using a different system). As can be seen,  $\Delta\langle r^2 \rangle$  has a clear  $Z$ -dependence that is probably due to the different rate of growth of deformation along each of the isotopic chains as  $N$  is reduced.

The next step in the development of laser spectroscopic techniques in nuclear physics is to combine the high resolving power of collinear spectroscopy of fast atomic beams with resonance laser photoionization. This is the aim of the Mainz-CERN-Troitsk collaboration.<sup>36</sup> It should result in further advances in the study of the properties of short-lived exotic nuclei that have unusual proton-to-neutron ratios, and lie well away from the stability band (see Fig. 2),<sup>37,38</sup> so that they are available in exceedingly small amounts. It is also important to note that the energy  $E_{\text{nu}}$  of an excited nucleus can be determined at least in principle, by measuring the negligible "isomeric" mass shift of the spectral line, due to the greater mass of the excited nucleus<sup>39</sup> (the mass increase is  $\Delta M = E_{\text{nu}}/c^2$ ). For atoms, the increase in mass is exceedingly small, but for vibrational transitions in molecules, it is actually observable. For example, in a diatomic molecule AB, the excitation of the nucleus of atom A produces a change in the vibrational frequency  $\nu_0$  by the amount

$$\frac{\Delta\nu_{\text{is}}}{\nu_0} = -\frac{E_{\text{nu}}}{2c^2} \frac{M_{\text{B}}}{M_{\text{A}}(M_{\text{A}}+M_{\text{B}})}, \quad (6)$$

where  $M_{\text{A(B)}}$  is the mass of atom A (B) with the nucleus in the ground state. When  $E_{\text{nu}} = 0.3$  MeV, the isomeric shift in the vibrational spectrum of an atom with  $A \approx 100$  is  $\nu_{\text{is}}/\nu_0 \approx 2 \times 10^{-6}$ , i.e., of the same order as the Doppler width of the vibrational-rotational molecular line at normal temperature. As laser cooling and localization methods for both atoms<sup>40</sup> and molecules advance, high-precision measurements of the frequencies of vibrational-rotational molecular transitions in radioactive atoms will probably become possible. Once accurate measurements and interpretations of the hyperfine structure of such transitions (due to the spin of the excited nucleus) become available, it will be possible to obtain data on the "isomeric" mass shift, i.e., it will be possible to use optical techniques to measure the mass defect  $\Delta M$  of an excited nucleus and, hence, its excitation energy. This may well lead to a more accurate relationship between the energy and wavelength scales for optical and  $\gamma$ -ray ranges.

### 3. LASER DETECTION OF VERY RARE NUCLEI

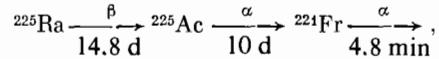
Accurate measurements of the structure and position of optical transitions, based on (1)–(5), yield information on nuclear parameters in ground and isomeric states. However, for many problems in nuclear physics and its applications, it is necessary to detect the nuclei themselves, i.e., to determine ultralow concentrations, down to single particles, including the atoms of rare elements (short-lived, transuranic, ultra-heavy, etc.), rare isotopes, nuclear isomers (energies, shapes, and densities), and so on. Laser methods of detecting single atoms,<sup>3</sup> especially the method of resonance photoionization,<sup>4</sup> seem to be very effective for the solution of such problems. As an illustration let us consider a few examples.

#### 3.1. Laser detection of rare atoms

We shall discuss this by considering the example of the atom of francium. This element (the isotope <sup>223</sup>Fr) was dis-

covered in 1939 in decays along the weak U channel. The amount of this longest-lived isotope (half-life 22 min) in 1 g of natural uranium is only  $6 \times 10^{-18}$  g. In the ISOLDE system at CERN, francium atoms can be produced by irradiating a uranium or thorium target with a proton beam. The first spectroscopic studies of francium<sup>41</sup> on this system were performed by French scientists who determined the wavelengths corresponding to transitions from the ground state to the four lowest-lying states<sup>42</sup> by magnetic deflection of atoms in a beam (the atoms were oriented by resonant laser excitation).

The Institute of Spectroscopy of the USSR Academy of Sciences has developed techniques for the detection<sup>43</sup> and spectral analysis of Rydberg states<sup>42</sup> of <sup>221</sup>Fr by resonance laser photoionization of francium fluxes that are lower by a factor of  $10^5$  than in the experiments on ISOLDE at CERN. The experiments with francium atoms were performed with a specimen containing about  $10^9$  atoms of <sup>225</sup>Ra, implanted to a depth of up to 100 Å in a tantalum foil. The specimen was prepared by collecting <sup>225</sup>Ra recoil nuclei from the radioactive decay of <sup>229</sup>Th, deposited in the form of a thin layer on the tantalum foil. The radioactive decay of <sup>225</sup>Ra atoms created the chain



containing <sup>221</sup>Fr. The stationary amount of <sup>221</sup>Fr in the specimen did not exceed  $3 \times 10^5$  atoms, and the rate of production was not more than  $1000 \text{ s}^{-1}$ . Since this experiment is probably the most sensitive in the history of optical spectroscopy, and has produced new spectroscopic data, it is opportune to consider it in greater detail.

The experiment on the laser detection of an atom is illustrated schematically in Fig. 5. As in the case of the nuclear/laser complex used by the Konstantinov Institute and the Institute of Spectroscopy collaboration (see Fig. 3), this experiment employed dye lasers with high pulse repetition frequency. Resonance ionization was produced by a two-step scheme involving the excitation of a Rydberg state which was then ionized by an electric-field pulse. However, in contrast to the other experiments, resonance ionization of the atoms to be detected was performed not in a beam, but in a heated cavity. The two-color laser radiation was introduced through an aperture in the cavity, and produced resonance excitation of the francium atoms to the Rydberg states. The electric-field pulse ionized the excited atoms and extracted the resulting ions through the aperture and into a time-of-flight ion-mass analyzer and a detector. The significant point was that francium atoms undergoing random motion within the cavity crossed the illuminated region several times, which increased the probability of their capture by the laser beam and, consequently, their ionization efficiency. The francium atoms were identified by the photoion resonance peak at 718.0 nm when the laser frequency was tuned to the first step, and by tuning the time-of-flight analyzer to the <sup>221</sup>Fr peak. The spectrum of Rydberg states was determined by varying the laser wavelength in the range 510–520 nm for a fixed wavelength corresponding to the first step.

The successful outcome of these experiments<sup>43,44</sup> confirms the proposal<sup>45</sup> that resonance photoionization could be used for the detection and analysis of single atoms of rare radioactive elements. It became clear that there was an alternative spectroscopy of elements that are not available in na-

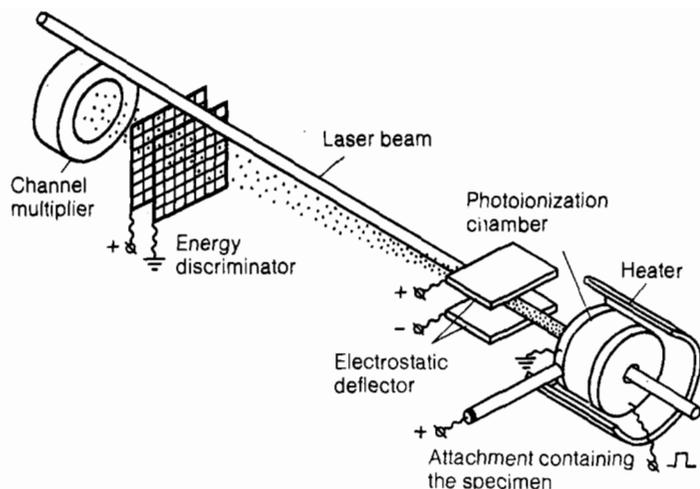


FIG. 5. Principle of an experiment on the photoionization detection and spectroscopy of the very rare francium atoms (from Ref. 43).

ture in amounts sufficient for optical studies. Instead of using expensive accelerator techniques to generate high fluxes of such elements,<sup>41,42</sup> it is possible to use weakly-radioactive sources of the elements (with activities less than  $10^{-6}$  Ci) in combination with the ultrasensitive laser photoionization spectroscopy that is now available to physics laboratories. This procedure can be used to detect rare radioactive atoms, including francium, even under natural conditions.

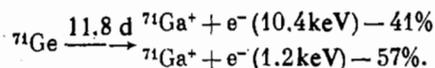
### 3.2. Laser detection of rare isotopes in the radiochemical neutrino detector

The numerous problems encountered in experimental nuclear physics and elementary-particle physics, in which it is necessary to detect exceedingly rare atoms in the environment of other elements, constitute a natural application for laser methods of detecting single atoms. For example, one possibility that has been discussed is the use of resonance laser photoionization in the detection of single atoms containing an excited (isomeric) nucleus, ultraheavy (transuranic) atoms,<sup>45</sup> ultradense nuclei,<sup>46</sup> nuclear shape isomers, including<sup>47</sup>  $^{240}\text{Am}$ , nuclei produced as a result of neutrino capture,<sup>48</sup> and so on. All experiments of this type are still at the discussion stage, but attempts to implement them have begun. Substantial advances may therefore be expected in the development of laser methods for the detection of single atoms in these interesting applications. We shall therefore confine ourselves to a brief discussion of one of the most obvious and intensively developing applications, namely, radiochemical detectors of solar neutrinos, capable of counting single atoms.

The best known is the Cl-Ar neutrino detector which is sensitive to high-energy neutrinos that form a relatively small fraction of the overall solar neutrino flux. This type of radiochemical neutrino detector was employed in the very accurate experiments using 600 tons of  $\text{C}_2\text{Cl}_4$  in a mine 1.6 km deep.<sup>49,50</sup> The single atoms of  $^{37}\text{Ar}$  that were produced in the target undergo  $\beta$ -decay with a half-life of 35 d, so that, under stationary conditions, the 400 m<sup>3</sup> of the target contain only 18 atoms of  $^{37}\text{Ar}$ . All these atoms can be extracted in 20 h, and can be collected in a miniature proportional counter containing pure argon with a few atoms of  $^{37}\text{Ar}$ . The  $\beta$ -decay of  $^{37}\text{Ar}$  produces Auger electrons with energies of 2.8 keV which are recorded by the proportional counter against a

very low background. The total efficiency of extraction and detection of the  $^{37}\text{Ar}$  atoms is 40–50%. This is the most sensitive radiochemical experiment that has ever been carried out. The measured rate of production of  $^{37}\text{Ar}$  in the target is  $0.39 \pm 0.05$  atoms per day, which is lower by a factor of about 3–4 than the standard model prediction.<sup>51</sup> This discrepancy is referred to as the “solar neutrino anomaly,” and has served as a stimulus to the development both of new theoretical models (see, for example, Ref. 52) and of other radiochemical detectors.

A proposal<sup>53</sup> has been made for a Ga detector of solar neutrinos, based on the  $^{71}\text{Ga}(\nu_e, e^-)^{71}\text{Ge}$  reaction, which makes use of the 39.6% of  $^{71}\text{Ga}$  in the natural isotopic mixture of  $^{69}\text{Ga}$  and  $^{71}\text{Ga}$ . One  $\nu_e$  capture per day takes place in the 50 tons of Ga. The half-life of  $^{71}\text{Ge}$  is 11.8 d, so that steady-state concentration of  $^{71}\text{Ge}$  in the 50 tons of Ga is only 17 atoms. The radiochemical Ga-Ge detector can operate on the same principle as the Cl-Ar detector: (1) chemical extraction of  $^{71}\text{Ge}$  and (2) detection of  $^{71}\text{Ge}$  decays in a Davis proportional counter.<sup>54</sup> The radioactive decays occur along two channels with the emission of Auger electrons:



Decays in the first channel can be detected in the Davis counter, but those in the second channel involve the production of low-energy electrons and are much more difficult to detect because of the background. If they are neglected, then the size of the Ga target that is necessary to produce a given signal-to-noise ratio must be increased by a factor of 2.5, which is very expensive. A method has been proposed for obviating this difficulty. It is based on discrimination against events with the formation of low-energy electrons, using the simultaneous laser detection of the Ga<sup>55</sup> atom. The suggestion is that the extracted Ge can be completely freed from Ga in the original target.

A radiochemical  $^{81}\text{Br}$ - $^{81}\text{Kr}$  solar neutrino detector with a threshold energy of 470 keV is being developed and, potentially, has the highest sensitivity to the neutrino flux from  $^7\text{Be}$  (Ref. 56). The  $^{81}\text{Kr}$  isotope produced in this system has a very long half-life ( $2 \times 10^5$  y), but it cannot be detected by a proportional counter. The natural resolution of this problem is to use resonance laser photoionization to

count single atoms of  $^{81}\text{Kr}$  (Ref. 57). For the same target dimensions as in the case of the Cl-Ar detector, it is expected that about two atoms of  $^{81}\text{Kr}$  will be produced per day in the case of compounds such as  $\text{CHBr}_3$ ,  $\text{CH}_2\text{Br}_2$ , and so on. A six-month exposure should give a few hundred atoms of  $^{81}\text{Kr}$  which can then be extracted by the same technique as in the Cl-Ar detector. Laser detection of the  $^{81}\text{Kr}$  isotopes can be achieved by combining resonance ionization of Kr atoms ( $Z$  selectivity) with mass separation of the photoions ( $A$  selectivity). Preliminary experiments involving the detection of 1 000 atoms of Kr by photoionization (Ref. 58) have led to the conclusion that the radiochemical/laser  $^{81}\text{Br}$ - $^{81}\text{Kr}$  detector of solar neutrinos is feasible.<sup>59</sup>

Another possibility that is being discussed involves the georadiochemical detection of solar neutrinos from  $^8\text{B}$ , using the  $^{97,98}\text{Mo}(\nu, e^-)^{97,98}\text{Tc}$  reaction.<sup>60</sup> The isotopes  $^{97}\text{Tc}$  and  $^{98}\text{Tc}$  produced in this reaction have a long half-life ( $2.6 \times 10^6$  y and  $4.2 \times 10^6$  y, respectively) and can also be detected by the highly selective stepwise photoionization technique.<sup>61</sup>

### 3.3. Laser detection of rare cosmogenic isotopes

There is a relatively large number of rare long-lived radioactive isotopes of cosmogenic origin, including those produced in nuclear reactions induced by cosmic rays in the upper air. They include  $^{10}\text{Be}$  from nuclear reactions induced by galactic cosmic rays on N and O nuclei,  $^{14}\text{C}$  from reactions induced by secondary neutrons in N, and  $^{26}\text{Al}$  from reactions on Ar. These isotopes are formed in the upper atmosphere and precipitate and accumulate on the Earth's surface and at the bottom of the oceans. The best known among them is radiocarbon,  $^{14}\text{C}$ , which is used in dating organic objects.<sup>62</sup> The isotope  $^{14}\text{C}$  with relative concentrations in the range  $10^{-12}$ – $10^{-15}$  can be used in radiocarbon dating objects and events in time intervals of 5 000–50 000 y (the half-life of  $^{14}\text{C}$  is 5730 y).

There are two universal methods at present for the detection of cosmogenic radioactive isotopes that are present in small concentrations. The first and most widely used method involves measurements of the specific activity of the specimen and subsequent comparison with the specific activity of a sample of zero age. The nuclear method suffers from the disadvantage that long-lived isotopes are observed in extremely rare radioactive-decay events. This means that to obtain an appreciable signal in an acceptable interval of time (a few days), it is essential to have a large number of rare radioactive isotopes in the specimen. The second method involves the use of an accelerator as an ultrasensitive mass spectrometer.<sup>63</sup> When this method is employed, and it does not involve the detection of radioactive decay events, the specimens can be much smaller but there are difficulties connected with the presence of abundant isotopes and isobars. The shortcomings of the two existing methods present an interesting challenge to laser methods. The problem is very complicated but basically solvable: it involves the detection of a few rare isotopic atoms against the background of  $10^{10}$ – $10^{20}$  other isotopes of the same atom.<sup>64–65</sup>

In principle, any of the techniques of laser spectroscopy that are capable of detecting single atoms<sup>3</sup> can be used for the highly selective detection of rare isotopes. The basic difficulty here is the attainment of maximum detection selectivity  $S$ , i.e., the detection of a small number  $N_A$  of the atoms of

a rare isotope A in the presence of a much larger number  $N_B$  of the main or background atoms B:

$$S = N_B N_A^{-1}. \quad (7)$$

Selectivity is due to the small isotopic shift  $\Delta\nu_{is} = \nu_A - \nu_B$  of the spectral line resulting from one or several successive resonance transitions in the atom from ground to excited states. The width of the spectral line is the natural limitation on selectivity because of the overlap between the wings of the closely-spaced spectral lines of atoms A and B. However, the nature of this limitation is significantly different for different methods.

The most promising methods are based on resonance multistep excitation of the rare isotope in a multifrequency laser field, using the isotopic shift of several successive resonance transitions. The net result is that the selectivities  $S_k$  at the successive excitation and ionization steps are multiplied together.<sup>67</sup> The principle of this method is illustrated in Fig. 6. For example, in the case of three-step excitation, using the isotopic shift at each step, the total selectivity of optical excitation can reach<sup>68</sup>

$$S_{opt} = S_1 S_2 S_3. \quad (8)$$

For moderate values such as  $S_k \approx (\Delta\nu_{is}^{(k)}/\Gamma_k^{-1})^2 \approx 10^4$ – $10^6$ , where  $\Delta\nu_{is}^{(k)}$  is the isotopic shift and  $\Gamma_k$  the homogeneous half-width of the  $k$ th transition, the total ionization selectivity can be very high:  $S_{opt} \approx 10^{12}$ – $10^{15}$ . Practical implementation of the principle of multiplication of selectivities encounters the difficulty that, for the most interesting long-lived isotopes, it is difficult to find an upward sequence of transitions with appreciable isotopic shifts, since only the ground state of the atom has an appreciable shift.

A universal way of overcoming this difficulty, and transforming the method of stepwise ionization into a real method of detection of rare isotopes, was proposed in Ref. 69. The idea is to use collinear stepwise photoionization of a beam of accelerated atoms. The atoms are accelerated and neutralized, and this is accompanied by the bunching of the longitudinal velocities of the atoms and, hence, by the re-

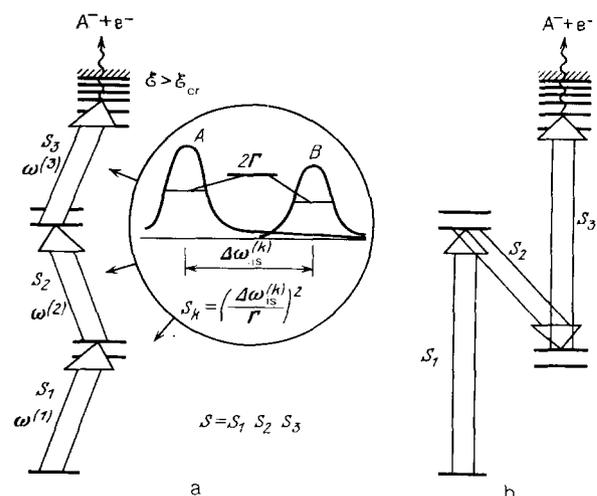


FIG. 6. Multiplication of selectivities during multistep excitation of a rare isotope A, with isotopically selective excitation at each step: a—successive upward excitation, b—successive excitation and de-excitation under the influence of laser radiation.

removal of Doppler broadening during collinear excitation.<sup>29</sup> The reduction in Doppler width is, in turn, accompanied by a Doppler shift of all the spectral transitions in accelerated atoms, which depends on the ion mass. This results in an artificial isotopic "mass" shift in all atomic transitions:

$$\frac{\Delta\nu_{is}}{\nu_0} = \frac{1}{2} (2eU)^{1/2} \left( \frac{1}{M_1^{1/2}} - \frac{1}{M_2^{1/2}} \right), \quad (9)$$

where  $M_i$  are the masses of the isotopes. When  $U = 10$  kV, this shift is greater by roughly an order of magnitude than the ordinary isotopic mass shift.

The first steps have been taken to develop this type of ionization laser detector of rare isotopes, using two-step collinear ionization of  $^{40}\text{K}$  atoms (0.012%) in a beam of accelerated potassium atoms. An ionization selectivity  $S \approx 10^5$  was achieved on one step. Further development of this method is continuing, using the multiplication of the selectivities of two or more steps, and it is intended to continue with much rarer cosmogenic isotopes.

#### 4. LASER ORIENTATION AND SEPARATION OF NUCLEI

There are many possible applications of selective laser excitation of atoms in which nuclei in particular states are produced for nuclear-physics experiments. This includes optical orientation (polarization) of nuclei in beams and targets, laser monochromatization (velocity selection) of protons in a beam, laser separation of isotopic and isomeric nuclei, and so on. These applications exploit the hyperfine interaction between nuclei and electrons, the isotopic shifts of electron states, and the Doppler effect in absorption spectra. Although they are all still in the development stage, rapid advances in laser technology suggest that we may realistically expect the implementation of these new techniques in nuclear-physics experiments in the near future. Let us briefly examine these possibilities.

##### 4.1. Laser orientation of nuclei

Orientation (or polarization) of atomic nuclei by circularly polarized optical radiation has been known since the pioneering work of Kastler.<sup>71</sup> The method relies on the absorption of polarized monochromatic radiation in a particular hyperfine transition. The interaction between the angular momentum of the electron and the spin of the nucleus ensures that the angular momentum of the absorbed photon is shared between them, and the result is that the nuclei in the ensemble of atoms become oriented in a particular direction (Fig. 7).

Optical orientation of nuclei had been successively used

before the advent of lasers in nuclear-physics experiments on the anisotropy of  $\beta$ -decay<sup>72</sup> and  $\gamma$ -decay<sup>73</sup> of oriented nuclei (the method employed was RADOP—Radioactive Detection in Optical Pumping). The possibilities of optical orientation of nuclei have become significantly greater since the advent of tunable lasers. This method can now be applied to any atoms with nuclear spin  $I > 0$ , including short-lived nuclei, either in a beam or in a dense gaseous target.

Laser orientation of excited nuclei in an atomic vapor has been investigated by a number of workers.<sup>74,75</sup> A particular feature of this method is that it can be used with short-lived nuclei (half-lives up to a few microseconds) that cannot be studied in a beam. Successful experiments have been carried out on the anisotropy of the  $\gamma$ -decay of the fissile isomer  $^{24m}\text{Na}$  (half-life 20 ms)<sup>75,76</sup> and the isomer  $^{134}\text{Ba}$  (lifetime 26  $\mu\text{s}$ ).<sup>75,76</sup>

An experiment has been carried out with a beam of laser-oriented nuclei in the case of the spontaneously fissile isomer  $^{240m}\text{Am}$  (lifetime of about  $10^{-3}$  s) in which an optical resonance with a transition in atoms containing the isomeric nucleus was detected by observing the anisotropy of fission fragments from oriented nuclei.<sup>77,78</sup> This technique has been used to measure the enormous isomeric shift (about 0.5  $\text{\AA}$ ) of the  $^{240m}\text{Am}$  line, and interesting data have been obtained on the large nuclear deformation of this isomer.

There is considerable interest in methods of producing polarized nuclei in beams and targets for studies of nuclear reactions that are sensitive to the orientation of the spin of the incident particles and of the target nucleus. This can also be done by laser orientation of lithium and sodium nuclei during resonant excitation by polarized radiation,<sup>79</sup> which has interesting applications in heavy-ion accelerators. However, there is even greater interest in the use of laser radiation to produce polarized beams of fast protons.

##### 4.2. Polarization and monochromatization of a fast proton beam

Here, there are several possibilities that rely either on indirect polarization in proton interactions in a polarized gas target, or on direct laser polarization of hydrogen atoms, followed by ionization (see Ref. 80).

The idea of producing a high-intensity beam of fast polarized protons in charge transfers between unpolarized protons and alkali-metal atoms, optically spin-oriented by laser radiation,<sup>81</sup> first emerged from the work of Zavoiskii<sup>82</sup> on the exchange of electron and nuclear polarization during charge transfer between fast protons and a polarized target.

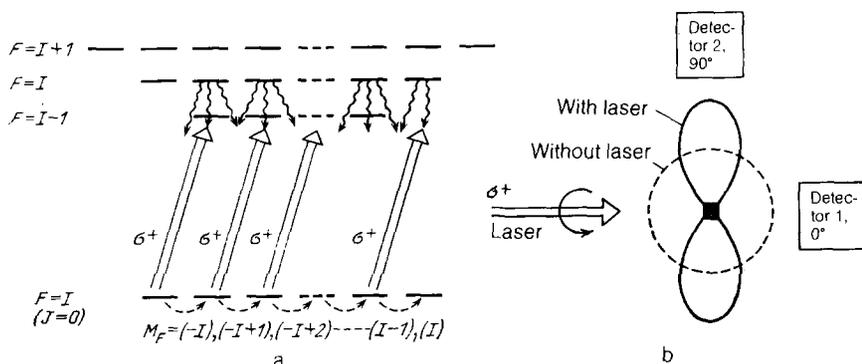


FIG. 7. Laser optical orientation of nuclei: a—optically pumped  $J = 0 \rightarrow J = 1$  transition with nuclear spin  $I = 1$ , leading to the population of the  $m_I = I$  sublevel as a result of the absorption of circularly-polarized radiation ( $\sigma^+$  transitions), b—appearance of anisotropic angular distribution of  $\gamma$ -rays from the E2 transition in oriented nuclei.

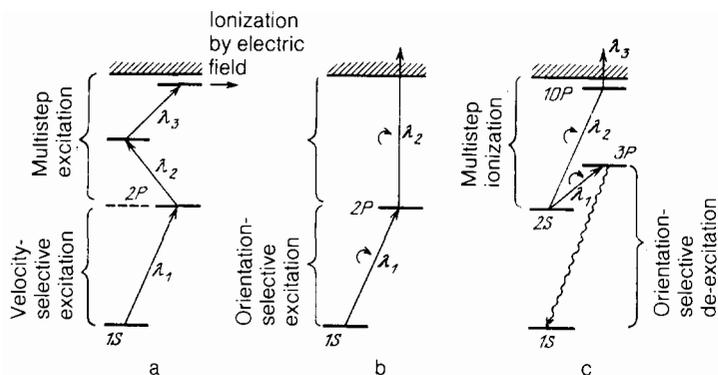


FIG. 8. Simplified methods of controlling the velocity (a) and polarization (b, c) of a proton beam, based on selective multistep photoionization of hydrogen atoms.

Successful experiments have been carried with a sodium charge-transfer target that ensures both a high degree of proton polarization (90%) and high charge transfer efficiency.<sup>80,83</sup>

Methods have also been suggested for producing highly monochromatic polarized beams of fast protons, based on orientational and (or) velocity selectivity of multiple-step ionization of fast hydrogen atoms by laser radiation.<sup>84</sup> Figure 8 is a simplified illustration of possible methods for the monochromatization<sup>85</sup> and polarization<sup>86</sup> of fast protons.

First, monochromatic laser radiation can be used to excite hydrogen atoms with a particular velocity component along a chosen direction (Fig. 8a). When the hydrogen atoms have relativistic velocities, and travel against the laser beam, the  $L_{\alpha}$  transition ( $\lambda_0 = 1215 \text{ \AA}$ ) can be excited in the visible range by visible radiation. Multistep photoionization of atoms in high-lying states can then be used to ensure that each excited hydrogen atom can be ionized with a quantum yield of about unity, i.e., a beam of protons can be produced with accurately defined velocity or energy (within a very narrow range). This is the principle of laser monochromatization of a proton beam.<sup>85</sup> To implement this principle, it is, of course, essential first to convert the initial beam of relativistic protons into hydrogen atoms. Modern tunable lasers and accelerators are, in principle, capable of producing a degree of monochromatization of about  $10^5$ . As the width of the energy spectrum of the proton beam is reduced, its intensity falls proportionally because the laser monochromator is a kind of "Maxwell demon" that selects hydrogen atoms with a particular velocity. However, the reduction in the beam intensity during monochromatization does not lead to the loss of the remaining unionized part of the beam energy distribution, which can be used in the accelerator in the usual way. We note that by varying the laser wavelength  $\lambda_1$  within the resonant 1S–2P transition, it is possible to isolate any particular narrow energy interval within the spectral width of the accelerated hydrogen atoms. Precise measurement of  $\lambda_1$  can then ensure the simultaneous absolute determination of the energy of the highly monochromatic protons.

Second, circularly polarized laser radiation can be used to excite a particular hyperfine transition (spin-selective or orientation-selective excitation) and then photoionize the selectivity-excited atoms (Fig. 8b). When proton polarization is produced at entry to the accelerator, so that the hydrogen atoms have nonrelativistic velocities, VUV laser radiation with  $\lambda(L_{\alpha}) = 1215 \text{ \AA}$  has to be used. The degree of polarization of protons under excitation via different hyper-

fine components of hydrogen<sup>88,89</sup> has been calculated, and the first model experiments have been carried out.<sup>90</sup> This method is, however, difficult to use to produce high-intensity, high-aperture beams, because of Doppler broadening. Another scheme has therefore been proposed in which hydrogen atoms are ionized from the metastable 2S-state (Fig. 8c). Here,  $H_{\alpha}$  laser radiation is first used to orient the nuclei by selective de-excitation of atoms from certain hyperfine sublevels, followed by multistep ionization of the oriented metastable atoms.<sup>86</sup> The basic difficulty in the practical implementation of this principle is the production of a sufficient number of hydrogen atoms in the metastable 2S-state. Polarization of accelerated protons can be achieved by using the relativistic Doppler effect and by polarizing relativistic hydrogen atoms via the 1S–2P transition by visible laser radiation,<sup>87</sup> i.e., in accordance with the scheme for the monochromatization of relativistic hydrogen atoms.<sup>85</sup>

It may therefore be expected that efficient methods of producing polarized, monochromatic, fast proton beams for accelerators will be developed. The success of these methods now depends mostly on advances in tunable lasers with parameters suitable for simultaneous operation with modern accelerators (long pulses, high pulse repetition frequency, the necessary wavelength, and so on).

#### 4.3. Laser separation of isotopes and nuclear isomers

The well-established methods for the laser separation of isotopes<sup>91,92</sup> are entirely suitable for producing multi-isotope beams and targets for nuclear-physics experiments, including searches for superheavy elements in heavy-ion accelerators. They are particularly valuable for rare isotopes for which separation by existing electromagnetic techniques is very expensive.

Some nuclear experiments require the availability of pure materials consisting of nuclear isomers. Selective laser photoionization of a mixture of atoms, containing nuclei in the ground and excited states, is practically the only way in which they can be separated. This is based on differences between the hyperfine structure of atomic transitions in excited and ground-state nuclei.<sup>93</sup> Laser separation of isomeric nuclei was discussed<sup>94</sup> as far back as 1973 in connection with, among other things, the production of a medium with inverted population of nuclear levels for a  $\gamma$ -ray laser<sup>95,96</sup> (see the reviews in Refs. 97 and 98). Selective laser photoionization of nuclear isomers was first observed in on-line experiments on the hyperfine structure and isotopic shift of the atomic lines of the radioactive isotopes of europium.<sup>93,99</sup>

The nuclear isomers of samarium-141g, 141m, and thulium-164g, 164m have recently been separated for the first time<sup>100</sup> by selective laser photoionization of atoms in an on-line experiment, using a proton accelerator and a mass separator of radioactive isotopes. The principle employed was similar to that illustrated in Fig. 3. Photoions with an excited nucleus of <sup>141m</sup>Sm or <sup>164m</sup>Tm, produced in isomer-selective three-step photoionization, were extracted by an electric field from the region of interaction between the laser radiation and the atomic beam, and were deposited on the cathode of a secondary-electron channel multiplier. Figure 9 shows the photoionization spectrum of a mixture of the <sup>141</sup>Sm isomers (laser wavelength tuned to the first step,  $\lambda_1 = 6004.18 \text{ \AA}$ ) together with an interpretation of the results. It is clear that the photoionization spectrum contains well-resolved lines belonging to <sup>141m</sup>Sm. This means that, when the laser frequency corresponding to the first step is *tuned in the range 1–4 GHz, a beam of photoions containing isomeric nuclei* is produced in the direction perpendicular to the atomic beam. This system can be used to investigate isomers with half-lives of the order of the time necessary to liberate them from the target (down to 1 s). A similar system, consisting of a mass separator, laser spectrometer, and photoionization chamber can be developed for other sources of isomeric nuclei, including beams of high-energy ions or a nuclear reactor.

### 5. MIXED NUCLEAR-OPTICAL $\gamma$ -TRANSITIONS

We now turn to converse effects, i.e., to manifestations of atomic-molecular structure in nuclear effects in the sim-

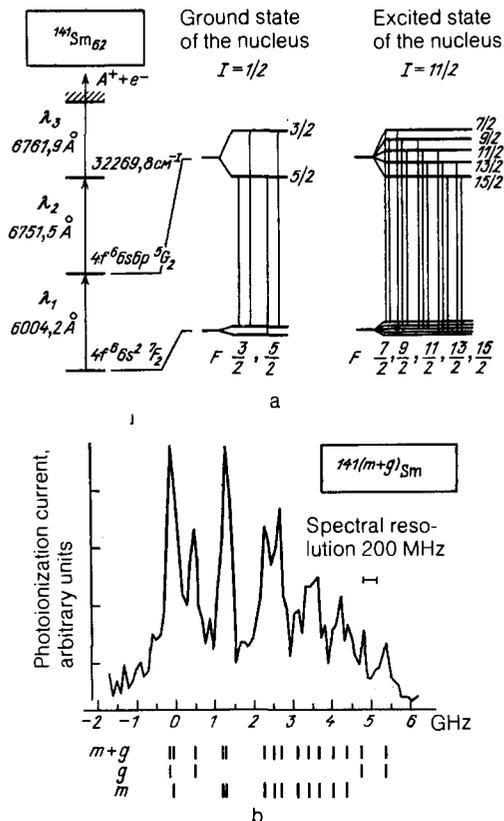


FIG. 9. Photoionization spectrum (bottom) due to the transition  $4f^6 6s^2 7F_2 \rightarrow 4f^6 6s 6p^3 G_2^0$  ( $\lambda_1 = 6004.18 \text{ \AA}$ ) in <sup>141</sup>Sm in the ground (*g*) and excited (*m*) nuclear states, produced by three-step photoionization of the atoms (top). The lowest diagrams show the position of the lines due to <sup>141</sup>Sm atoms with ground-state (*g*) and isomeric (*m*) nuclei (taken from Ref. 100).

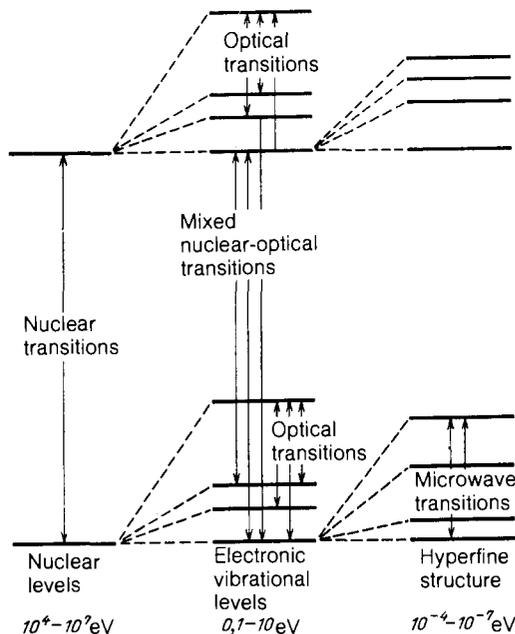


FIG. 10. Hierarchy of nuclear, optical, and hyperfine transitions in an atom and the appearance of mixed nuclear-optical transitions.

ple case of  $\gamma$ -transitions not in a bare nucleus, but in a nucleus surrounded by atomic electrons, or a nucleus in a molecule. The essence of this situation can be understood by considering the hierarchy of the energy levels of a nucleus in an atom (molecule), shown in Fig. 10 which is a development of Fig. 1. Figure 10 illustrates transitions occurring in the nucleus (nuclear spectroscopy), optical transitions in the atom and molecule (optical spectroscopy), and transitions between hyperfine components (microwave spectroscopy). The hyperfine interaction leads to the “mixing” of optical and “microwave” transitions, and to the appearance of the hyperfine structure of optical lines, discussed in the preceding Sections. In precisely the same way, it is possible to produce “mixing” of nuclear and optical transitions, and an optical structure in  $\gamma$ -lines. However, the interaction mechanism is now quite different, i.e., purely kinematic: it involves the recoil of the nucleus when the  $\gamma$ -ray is emitted, and the inertia of the electron shell or of other atoms in the molecule.

In the bare nucleus, the emission and absorption  $\gamma$ -ray lines are shifted to relative to each other by twice the recoil energy:

$$2R = \frac{E_0^2}{Mc^2}, \quad (10)$$

where  $M$  is the mass of the nucleus and  $E_0$  is the energy of the nuclear transition (we suppose that  $E_0 \ll Mc^2$ ). The shift of the emission or absorption line frequencies is due to the change in the  $\gamma$ -ray energy resulting from recoil. When the nucleus is in an atom or a molecule, the laws of conservation of momentum and energy allow a change not only in the translational state of the nucleus, but also in its internal state (electronic, vibrational, and rotational). The energy of the absorbed or emitted  $\gamma$ -ray is then given by:

$$\hbar\omega_\nu^\pm = E_0 \pm R + \hbar\mathbf{k}_\nu \cdot \mathbf{v}_0 \pm (\mathcal{E}_f - \mathcal{E}_i), \quad (11)$$

where  $\mathbf{v}_0$  is the velocity of the atom (molecule),  $\mathcal{E}_i$  and  $\mathcal{E}_f$

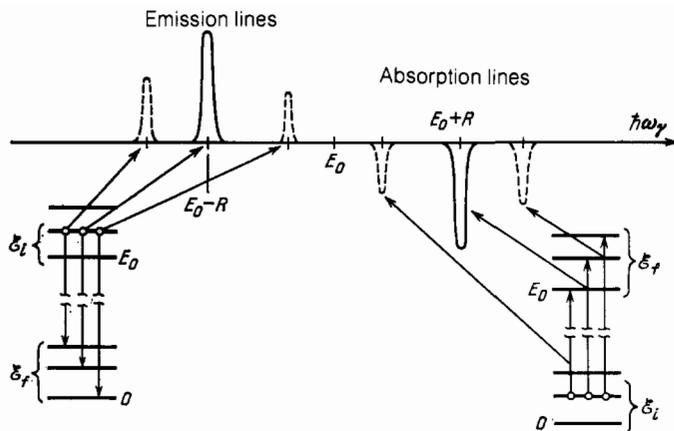


FIG. 11. Spectrum of nuclear  $\gamma$ -transitions in an excited atom (or molecule). Left— $\gamma$ -ray emission lines, right— $\gamma$ -ray absorption lines.

are, respectively, the initial and final internal energies, and the positive and negative signs correspond to the absorption and emission of a  $\gamma$ -ray, respectively. The first term represents the unshifted transition frequency (Mössbauer line of a nucleus in a crystal), the second represents the frequency shift due to the change in the translational state of the particle on recoil, the third represents the Doppler shift, and, finally, the last term represents the shift of the spectral line due to the change in the internal state of the atom (molecule) due to recoil. The last term describes the so-called mixed nuclear-optical transitions.

Figure 11 shows the absorption and emission  $\gamma$ -ray spectrum of a nucleus in an initially excited atom or molecule with  $\mathcal{E}_i > 0$ . When a  $\gamma$ -ray is emitted, part of the nuclear excitation energy can be transferred to the internal state of the particle ( $\mathcal{E}_f > \mathcal{E}_i$ ), which results in the appearance of a satellite shifted towards the "red" end relative to the transition energy  $E_0 - R$ , and the state of the particle does not change. Similarly, the excitation energy of the atom (molecule) can be transferred together with the excitation energy of the nucleus to the  $\gamma$ -ray, which produces a "blue" satellite of the  $\gamma$ -line relative to  $E_0 - R$ . A similar picture arises in  $\gamma$ -ray absorption.

Laser radiation can be used to change the population of atomic or molecular states and thus modify the spectrum of mixed  $\gamma$ -transitions in two ways: by changing the intensity of the corresponding satellites and by inducing new satellites that are shifted toward longer wavelengths relative to the  $\gamma$ -absorption line  $E_0 + R$  and toward shorter wavelengths relative to the emission line  $E_0 - R$  (Refs. 13 and 101). Since the excitation of atoms and molecules can be velocity-selective under excitation in a narrow interval of the Doppler-broadened absorption line, this enables us, at least in principle, to produce narrow resonances on Doppler-broadened  $\gamma$ -ray lines.<sup>102, 103</sup>

For a nucleus in a molecule, there are satellites<sup>13</sup> due to changes in the electronic, vibrational, and rotational states. The probability of electron-nuclear transitions in the molecule is very small because it contains the small parameter  $(m_e/M)^2$  where  $m_e$  is the electron mass and  $M$  the mass of the molecule.<sup>104</sup> We note that this enables us to assume that the dissociation of a molecule during the emission or absorption of a  $\gamma$ -ray (Szilard-Chalmers effect) occurs predominantly not as a result of the excitation of the electronic states of molecules, but as a result of the direct rupture of bonds

within the molecules in its electronic ground state. When a  $\gamma$ -active nucleus is external to the centre of mass of the molecule, strong rotational satellites of the  $\gamma$ -transition are found to arise, but their shift is small in comparison with  $R$  and the Doppler width. A more favorable situation occurs for the vibrational satellites because the mean energy expended in exciting the vibrations of the molecules in a  $\gamma$ -transition is:<sup>105</sup>

$$E_{\text{vib}} \approx \frac{1}{2} R \frac{M - M_\gamma}{M_\gamma}, \quad (12)$$

where  $M_\gamma$  is the mass of the  $\gamma$ -active nucleus and  $M$  is the mass of the entire molecule. When  $M_\gamma \ll M$ , the resulting vibrational satellites of the  $\gamma$ -transition exhibit a much greater shift than for the bare nucleus. In all cases, this type of a nuclear satellite of a molecule can only be observed for strong  $\gamma$ -ray sources, since the cross section for the nuclear transition is small and the situation is complicated by Doppler broadening.

Electronic satellites<sup>13</sup> appear in the case of a nucleus in an atom, but their intensity is again low because of the presence of the small parameter  $m_e/M$ . Simple qualitative considerations suggest that the deactivation of an isomeric nucleus may be accelerated by the transfer of part of its angular momentum to the electron shell, i.e., by the radioactive decay of the metastable nuclear excited state, accompanied by an electron-nuclear transition. However, calculations show that this effect is small.<sup>106</sup> The electronic satellites of  $\gamma$ -transitions are therefore also very difficult to observe. A different situation arises in the case of a nucleus in a multiply-charged ion<sup>107</sup> because the mechanism responsible for the appearance of satellites in a neutral atom and a multiply-charged ion is significantly different. In the atom (loose electron shell) the shell is "shaken" during the interaction between the  $\gamma$ -ray and the nucleus. In the multiply-charged ion (rigid electron shell), the  $\gamma$ -ray can interact directly with the electrons. This mechanism is significant for nuclear dipole transitions, and predominates at  $\gamma$ -ray energies below  $4Z^* \text{ keV}$ , where  $Z^*$  is the effective charge of the nucleus for an optical electron in the ion. The intensity of the electronic satellites is determined by the small parameter  $\mu_p^{2\lambda}$ , where  $\lambda$  is the multipolarity of the nuclear transition,  $\mu_p \approx Z/2$  is the relative mass of the proton (and not the electron!), and  $Z$  is the nuclear charge. Moreover, the electronic satellite shifts in multiply-charged ions are much greater than in the neutral

atom because the electron transition energies in the former are much greater. This means that the electronic satellites corresponding to the emission and absorption of a  $\gamma$ -ray by a nucleus in a multiply-charged ion are not only strong, but they are not overlapped by the Doppler profile of the  $\gamma$ -line.<sup>107</sup> We thus have a real possibility of observing them in high-temperature plasmas. It is possible that the electron-nuclear satellites will be of interest in relation to the  $\gamma$ -ray laser because they are easier to use to produce population inversion than the pure nuclear transitions.

## 6. OTHER TOPICS

Of course, we have not been able to cover all the topics that lie at the interesting interface between atomic and nuclear physics. They have become particularly active in recent years because of the availability of lasers and have probably stimulated advances in lasers themselves, especially in the short-wavelength range. There are two reasons for this. First, I wanted to concentrate my attention on a group of intimately related questions that can be more readily examined in a brief form. Second, it is precisely these questions that are being actively investigated at the Institute of Spectroscopy of the USSR Academy of Sciences in collaboration with other institutes and laboratories. However, it will be useful in conclusion to list other important directions of research in which lasers are being used, or will be used, in nuclear physics and elementary-particle physics.

(1) *The effect of the weak interaction between electrons and nuclei in atoms and molecules on parity violation due to neutral currents.* This problem is discussed in the monograph by Khriplovich.<sup>10</sup> Atoms exhibit a number of effects due to parity violation. First, atomic vapors exhibit weak optical activity<sup>11</sup> which has been detected in a series of well-known experiments on the rotation of the plane of polarization of laser radiation near atomic resonance absorption lines.<sup>108,109</sup> Second, atomic  $nS_{1/2}$  and  $nP_{1/2}$  states can mix, and this is observed, for example, as weak polarization of the fluorescence from an excited atom.<sup>110</sup> The effect has also been seen in experiments with the heavy cesium atom.<sup>111</sup> Parity violation in molecules should lead to a negligible splitting of the energy levels of left-handed and right-handed molecules,<sup>12</sup> but the magnitude of this is beyond the range of present-day experiments. On the other hand, rapid advances in deep cooling and trapping of atoms (and, eventually, of molecules too) by laser radiation open up completely new avenues for very precise experiments.

(2) *Hyperfine splitting of the energy levels of muonic atoms and molecules occurs in the optical range and can therefore be investigated by laser techniques.* So far, the only example of this is the measurement of the  $2S_{1/2} - 2P_{1/2}$  energy difference (Lamb shift) in muonic helium  ${}^4\text{He}\mu$ , which lies near 811.7 nm (Refs. 112 and 113). This experiment succeeded in verifying the contribution of vacuum polarization to this splitting with a precision of 0.001. It is planned to repeat this experiment with the latest laser technology.<sup>114</sup> Another important quantity that would be interesting to measure with infrared laser radiation is the hyperfine splitting in muonic hydrogen.<sup>115</sup> It may well be that laser radiation could be used to influence the rate of production of mesic molecules (see, for example Ref. 116). The development of strong sources of mesons in physics research centers with well established laser-physics facilities will lead to the

development of new experiments in this area.

(3) Finally, in the very long term, it may be possible to *use laser radiation to influence processes occurring in the nuclear interior.* Of course, the first candidate for this is the  $\beta$ -decay process which in itself is slightly sensitive to the chemical environment. For example, the possible effect of the ionization of radioactive atoms by powerful laser radiation on the rate of internal conversion and K-capture has been examined.<sup>117</sup> However, rigorous calculations<sup>14</sup> have shown that this requires very strong wave fields that have not as yet been produced. It seems that the effect of strong laser radiation is not observable even for forbidden beta-decays.<sup>118-119</sup>

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