Use of coherent-optics methods in nuclear-physics research

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The use of tunable lasers to obtain polarized and coherent ensembles of nuclei is discussed. The possibilities and advantages of laser technology in this field are demonstrated by various examples.

INTRODUCTION

In recent years, tunable lasers (dye lasers) have been increasingly used in experimental investigations into the properties of nuclei. In these lasers, the frequency of the laser radiation can be smoothly adjusted in the complete visible range of the spectrum. Lasers are characterized by a high energy resolution ($\Delta E/E = 10^{-8}$–10^{-9}) and a high power of the generated radiation (up to 1 W in the continuous-wave (cw) regime and up to 10^5 W in a pulse). It is possible to obtain laser radiation with a definite polarization, the degree of circular or linear polarization effectively reaching 100%. Such properties of lasers with tunable frequency open up new possibilities for investigating nuclear structure and make it possible to control the direction of the nuclear spin and establish a phase connection between nuclear substates. The study of nuclear processes taking place under extremal conditions of a strong electromagnetic field also becomes realistic.

At the present time, one can identify at least four groups of physics problems that can be solved by means of dye lasers.

1. The determination of the mean-square charge radii of nuclei and the spins and magnetic and quadrupole moments of the ground and isomer states of nuclei. The method is based on selective excitation of the hyperfine-structure components by a laser beam with tunable frequency. The effect is detected by fluorescence (optical methods) or by the anisotropy of the nuclear radiation. The method makes it possible to work with small amounts of a substance. Measurement of the hyperfine structure and the isotopic shift by laser spectroscopic methods is essentially a direct measurement of the shape and size of the charge distribution in the nucleus. This aspect of laser spectroscopic methods is very fundamental, since, in contrast to the existing methods of analyzing results in nuclear physics, it does not depend on model properties of the nuclei or decay channels of radioactive nuclides.

2. Orientation of nuclei by a laser beam. The acquisition of coherent ensembles of nuclei. Study of the decay of such systems. The investigation of fundamental interactions by means of such systems. Nuclear reactions on targets with coherent properties. Determination of the magnetic and quadrupole moments of the ground and isomer states of nuclei.

The method is based on the excitation of a number of hyperfine-structure components by polarized laser radiation, this resulting in polarization and coherence in the atomic shell. Polarization of nuclei and nuclear coherence are obtained through the hyperfine interaction. The method is highly selective and makes it possible to measure high degrees of polarization (up to 100%) and appreciable coherence ($\sim$11%).

3. The search for and detection of rare events: superheavy and superdense nuclei, double β decay, neutrino experiments, etc.[1]

The method is based on selective ionization of atoms with the studied nuclei and subsequent study of the ion in ion traps. According to the existing estimates, the laser method of rare-event detection could become one of the most sensitive methods.

4. Nuclei and nuclear reactions in a field of strong electromagnetic radiation. There is a hope of finding new, previously unknown effects, since hitherto the behavior of nuclei in a field of strong electromagnetic radiation has been little studied.[2]

In this review, we present results of investigations in the field of application of tunable lasers to obtain and study polarized and coherent ensembles of nuclei. The possibilities and advantages of laser technology in this field are demonstrated.

Optical methods of polarizing nuclei and creating coherent ensembles were initiated by the classical studies of Kastler[3] and Hanle.[4] In recent years, the methods of high-resolution laser spectroscopy have been widely used to investigate isotopic shifts and the hyperfine structure of atoms with short-lived nuclei. The methods of coherent optics have been used much less often in nuclear physics. Nevertheless, the development of these methods at the present time, in which pulsed and cw dye lasers are available, offers the prospect of a significant extension of the field of application of optical methods in nuclear physics. In the present review, we consider the following questions:

1. Methods of orienting and aligning nuclei by means of laser radiation with detection through optical and nuclear radiations.

2. Coherent effects in the electron shell, investigation of the Hanle effect and level crossing for atoms with radioactive nuclei, and the determination of the quadrupole moments of nuclei.

3. The acquisition of coherent ensembles of nuclei (namely, ensembles in which nuclear substates differing in their magnetic quantum numbers are connected in phase) and investigation of the effects of nuclear coherence in optical and nuclear radiations.

We shall not consider in detail questions relating to the traditional methods of high-resolution laser spectroscopy employed to investigate the properties of short-lived nuclei. Several reviews have been devoted to this question.[5–7]

ORIENTATION OF NUCLEI BY OPTICAL PUMPING IN THE ATOMIC GROUND STATE

Investigation of the decay of oriented nuclei makes it possible to obtain a variety of information about both the

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characteristics of nuclear states and the properties of fundamental nuclear interactions.[8] However, the wide use of these methods comes up against serious difficulties and limitations in the currently developed methods for orienting nuclei. A significant extension of the ambit of nuclear-physics research can be achieved by the development of reliable optical methods for orienting nuclei.

The possibility of orienting nuclei by means of optical pumping of atoms with circularly polarized light was already noted by Kastler,[3] but the first attempts to use this method in nuclear physics date from the middle of the seventies. The most work was done on the orientation of the electron shell of atoms by means of circularly polarized light. A review of the work done with the aim of selecting the optimal conditions for optical pumping of atoms, relaxation times, etc., can be found in Ref. 9. All these studies were made with flashlamp light sources. Some aspects of the optical pumping of atoms by means of laser radiation are discussed in Refs. 10 and 17. It is natural that the orientation of the atoms and the degree of orientation were also established by means of optical methods or from the deflection of polarized beams of atoms in inhomogeneous magnetic fields. In nuclear physics, one can use the fact that the existence of the hyperfine interaction between the nucleus and the electron shell leads to orientation of the nuclear spins if the atomic spins are oriented. Dynamical orientation of a nucleus in this process comes about through the preferred population of one of the magnetic sublevels of the atom.

The first studies on the optical orientation of radioactive nuclei were made by Otten's group.[11] A fundamental feature of these studies was the detection of the nuclear orientation through the anisotropy of the nuclear β or γ radiation ($\beta-\gamma=$ RADOP). These studies can be divided into two groups. The first group[12,18] used mercury isotopes. The optical pumping was done by means of a mercury lamp, the scanning of the wavelength of the light was realized by a magnetic field, and the mercury vapor was created by heating a collector onto which an ion beam was precipitated after an isotope separator. The use of such a scheme to investigate other elements involves serious difficulties due to the differences of the relaxation effects and chemical activity. The second group of studies[11,13] related to the orientation of short-lived isotopes of Na and K formed directly in reactions with protons or deuterons on Ne and Ar gas targets. In this special case, one can avoid the main methodological difficulty of working with radioactive isotopes, namely, the preparation of a sample in a cell in the necessary chemical state with minimal amount on the cell walls; however, it is almost impossible to work with long-lived isotopes because of the small yield of them in the reaction.

The Na and K isotopes were oriented by spin-exchange collisions with Rb vapor optically oriented by means of special lamps. The nuclear-physics experiments made in Refs. 11–13 had a number of shortcomings.

1. The asymmetry of the nuclear radiation was measured by comparing counts at only two angles, since detectors with large solid angles were used.

2. The work with optically oriented samples was demonstrated only in some special cases.[11–13]

The main difficulty was that already in the preparation of the sample an appreciable portion of the employed substance was precipitated on the walls or formed different chemical compounds, so that a background was created that made it impossible to measure the asymmetry of the nuclear radiation.

3. The optical pumping was achieved by means of flashlamp sources, and this greatly restricted the experimental possibilities.

In Refs. 14–16 a fairly universal apparatus and method of orienting stable ^{23}Na nuclei as well as the long-lived radioactive ^{22}Na and ^{24}Na nuclei by means of a dye laser are described. Some of the difficulties in preparing the samples can be overcome by means of pulsed laser evaporation of matter. The degree of orientation of the nuclei was estimated through the angular distribution and polarization of the fluorescence from nuclear transitions and nuclear γ decay.

Mechanisms for orienting atoms and nuclei by optical pumping

Nuclei are oriented in the process of optical pumping by the preferred population of certain Zeeman sublevels in the atom. There are two mechanisms of optical pumping[9]: *depopulating* and *repopulating*. In the first, atoms accumulate in the Zeeman sublevels that more weakly absorb the optical radiation; in the second, there is preferred population of the sublevels to which transitions from an excited state take place most strongly. In what follows, these two forms of pumping will simply be called optical pumping, with the particular form being identified where necessary.

For orientation of nuclei by optical pumping there must be a hyperfine interaction between the nucleus and the electron shell. This interaction leads to the appearance of the hyperfine structure in the spectra of atomic transitions. In the atomic ground state, orientation of the nucleus can occur in the following three cases:

(1) there is a hyperfine interaction in the atomic ground state;

(2) there is a hyperfine interaction in the excited atomic state;

(3) there is a hyperfine interaction in both the ground and excited atomic states. When we say that "there is a hyperfine interaction" we mean that the following conditions are satisfied:

(1) in the excited state

$$\tau_{\text{hpf}} \lesssim \tau_{\text{sp}}, \quad \tau_{\text{hpf}} \lesssim \tau_{\text{st}};$$

(2) in the ground state

$$\tau_{\text{hpf}} \lesssim \tau_{\text{st}},$$

where $\tau_{\text{hpf}} \sim h/\Delta E_{\text{hpf}}$ is the characteristic time of the oscillations in the wave function of the atom associated with the interaction of the nucleus with the electron shell, ΔE_{hpf} is the energy of the hyperfine interaction (the hyperfine splitting), τ_{sp} is the spontaneous-decay time of the atomic level, and τ_{st} is the stimulated-transition time, i.e., the time during which the atomic sublevel is populated in the presence of optical excitation.

If hyperfine interaction is absent in both the ground and excited atomic states, then nuclear orientation by optical pumping will not occur.

If matter vapor is illuminated by polarized laser radiation at the frequency of an atomic transition under the condition $\tau_{\text{st}} \sim \tau_{\text{hpf}}$, then there is excitation of the atoms to the

fine-structure states that during the time τ_{hpf} decay to the hyperfine-structure levels. One can speak of a hyperfine transition being distinguished and a nucleus being oriented directly during the time of excitation of the atomic level only if the time scale satisfies $t \gtrsim \tau_{hpf}$. In the present review, we consider processes in precisely this time interval. As an example, we give the characteristic times and corresponding energy widths for ^{23}Na. The lifetime of the first excited atomic levels $P_{1/2}$ and $P_{3/2}$ is $\tau_{sp} \sim 1.6 \times 10^{-8}$ sec, to which there corresponds the intrinsic width $\Delta E = 9.76$ MHz. The hyperfine splitting in the $P_{1/2}$ state is $\Delta E = 190$ MHz, to which there corresponds the time $\tau_{hpf} \sim 8 \times 10^{-10}$ sec, while in the $S_{1/2}$ ground state $\Delta E_{hpf} \sim 1.7$ GHz; then $\tau_{hpf} \sim 9 \times 10^{-11}$ sec. Thus, in studying optical pumping in the D_1 line (excitation of the $P_{1/2}$ state) in ^{23}Na on a time scale $t \gtrsim 8 \times 10^{-10}$ sec one can ignore the oscillations in the wave function of the atom due to the hyperfine interaction.

In the process of optical pumping, not only the total spin of the atom, **F**, but also the spin of the nucleus, **I**, where $\mathbf{F} = \mathbf{I} + \mathbf{J}$, and **J** is the spin of the electron shell, is oriented. The orientation of the spins takes place in the atomic ground state. The elements for which orientation of the nuclear spin by optical pumping is possible can be divided into two classes:

(1) elements for which there is a hyperfine splitting in the atomic ground state;

(2) elements for which there is no hyperfine splitting in the ground state.

In the second case, it is important that the nucleus has been oriented in an atomic excited state, i.e., after absorption of polarized laser radiation, and this, in its turn, imposes a number of additional requirements on the experiment, which reduce to suppression of relaxation processes in the atomic excited state.

There are also a number of requirements common to classes 1 and 2. By optical pumping one can orient substances in the gaseous state with the S configuration of the electron shell, since otherwise the relaxation processes are, as a rule, so strong that the effects of the orientation become experimentally unobservable.[18] In addition, it is necessary to create conditions under which the presence of atoms on the walls of the cell does not make a significant contribution to the observed effect. This, in its turn, reduces the number of nuclei that can be oriented.[16]

Up to the present time, orientation has been achieved for the nuclei of alkali elements in the atomic state and a number of ions of other elements having a similar electron configuration (class 1).[9,11,13] For class 2, the nuclei of helium, cadmium, barium, ytterbium, americium, and lutecium have been oriented (Refs. 9, 12, 19, and 20). In addition, polarization has been obtained in metastable states of a number of noble gases and in lead and thallium.[9]

Physical processes associated with orientation of alkali elements

We consider the orientation of nuclei by optical pumping for the example of alkali atoms. The mechanism of the optical pumping for these atoms and for a number of ions with the $S_{1/2}$ electron configuration of the ground state is the same, although in each particular case there are specific technical difficulties in the implementation.[9]

The level scheme of the atoms of alkali elements is shown in Fig. 1. In what follows, we shall use this notation: **F** is the total spin of the atom, **I** is the spin of the nucleus, **J** is the spin of the electron shell, **S** is the spin of a photon, and m_j is the projection of the spin **J** onto the direction of the laser radiation. Repopulating and depopulating pumping are equally effective for atoms of the alkali elements. The photon absorption operators for the D_1 and D_2 lines have the form[9]

$$\delta\Gamma_1 = R_1 (1 - 2\mathbf{J}\mathbf{S}_1); \ \delta\hat{\Gamma}_2 = R_2 (1 + \mathbf{J}\mathbf{S}_2), \qquad (1)$$

where R_1 and R_2 are the mean rates of absorption in each of the lines, and $\mathbf{S}_1$ and $\mathbf{S}_2$ are the spins of the photons responsible for the excitation of the $P_{1/2}$ (D_1 line) and $P_{3/2}$ (D_2 line) atomic levels. The absorption operator for light in which both D components are present has the form

$$\delta\hat{\Gamma} = (R_1 + R_2) - (2R_1\mathbf{S}_1 - R_2\mathbf{S}_2)\,\mathbf{J}. \qquad (2)$$

When the D_1 and D_2 components in the irradiating light have the same polarization, i.e., $S_1 = S_2$, and their intensities are approximately equal [in this case $2R_1 \approx R_2$ (Ref. 9)], it can be seen from (2) that polarization of the atom and nucleus does not occur. This is a consequence of a more general sum rule[9] from which it follows that if when an unpolarized atom is excited the transitions from the S state of the electron shell take place with equal intensity to all components of a fine-structure multiplet, then the population of the Zeeman components of the S state after the spontaneous decay will be uniform. Thus, to achieve a nonuniform population of the Zeeman sublevels in the S state, it is necessary to choose the spectral composition of the irradiating light in such a way that the transitions in the multiplet are excited with different probabilities. Using both D lines, one can obtain an appreciable degree of orientation of the atom and the nucleus only if the spins $\mathbf{S}_1$ and $\mathbf{S}_2$ are antiparallel. Frequently, only one D line is used, and in this case one can also obtain an appreciable degree of orientation.

For alkali elements, the spin of the electron shell in the $S_{1/2}$ ground state is $J = 1/2$. The total spin **F** of the atom takes the two values $I \pm 1/2$. A circularly polarized photon σ^+ has angular-momentum projection $+1$. The resonance absorption of such photons by atoms will take place in accordance with the selection rules $\Delta\mathbf{F} = 0, \pm 1$ for the total angular momentum and $\Delta m_F = +1$ for its projection. In the case of multiple absorption of σ^+ photons and subsequent decay, atoms accumulate in the ground state in the magnetic sublevel with the maximal value of the projection of the total angular momentum **F** onto the direction of the laser beam,

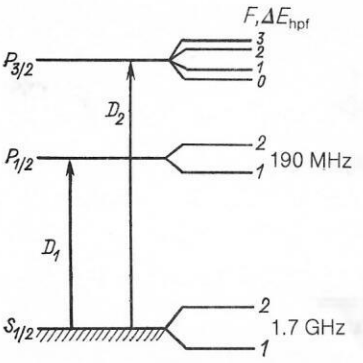

FIG. 1. Level scheme of the ^{23}Na atom, $I = 3/2$.

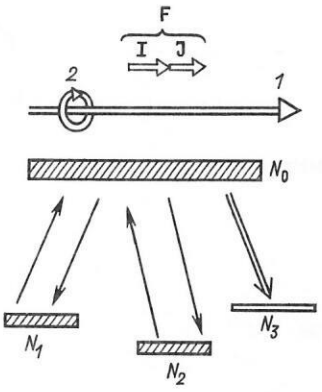

FIG. 2. Groups of states of atoms of alkali elements that participate in the optical pumping process: (1) laser beam; (2) direction of polarization of the laser radiation.

i.e., there is orientation of the spin of the atom and the nucleus. Figure 2 shows the groups of states that participate in the optical pumping. Here, N_1 are the sublevels of the ground-state doublet with $F = I - 1/2$, and N_2 are the same for $F = I + 1/2$ except for the sublevel with maximal projection $m_F = I + 1/2$, which is denoted by N_3; N_0 are the doublet states of the excited $P_{1/2}$ level with $F = I \pm 1/2$. In accordance with the selection rules, absorption takes place from the groups of levels N_1 and N_2, but not from N_3. However, decay takes place to not only the N_1 and N_2 states but also the N_3 state. In the case of multiple excitations and decays, atoms accumulate in the state N_3, i.e., they are oriented. This means that the nuclear spins are also oriented, since in the N_3 state the direction of the nuclear spin coincides with the direction of the atomic spin. The time during which an appreciable degree of orientation (up to 100%) of the atoms and nuclei occurs depends on the power of the exciting radiation and on the probabilities of the transitions between the states in which we are interested; the time is related to the lifetime of the excited atomic level and is limited by it.

By means of nonlaser light sources an orientation time of atoms and nuclei during 10^{-2}–10^{-3} sec could be ensured in the best case; moreover, the frequency of the pumping light remained fixed or could be changed only in a small range. The spectral resolution was low, and this made it difficult to measure the hyperfine splitting in the atom and, hence, to determine the quadrupole and magnetic moments of the nuclei. Dye lasers, operating in both the cw and the pulsed regime, permit excitation of an atomic transition during a time much shorter than the lifetime of the excited atomic state (about 10^{-8} sec), i.e., the speed with which nuclei can be oriented by optical pumping is now limited only by the lifetime of the excited atomic state. The wavelength of the laser radiation can be varied in a fairly wide range, and this, in its turn, makes it possible to carry out experiments with many elements with excitation of different atomic transitions. The highly monoenergetic nature of laser radiation, which can reach several megahertz, makes possible pumping in different components of the hyperfine structure of an atom and the determination with high accuracy of the quadrupole and magnetic moments of nuclei.

For the orientation of the nuclei, one can use either pulsed or cw dye lasers. At the present time, higher powers (in a pulse) of the laser radiation have been achieved in pulsed lasers, and this makes it possible to excite atomic transitions in shorter times than when cw lasers are used. When cw sources of polarized pumping radiation are used, the maximal possible degrees of polarization (reaching 100%) can be achieved. If pulsed lasers are used, one can work with ensembles of atoms and nuclei in which orientation has occurred after one, two, etc., cycles of excitation of an atomic transition and spontaneous decay to the ground state. It should be noted that the orientation of nuclei by pulsed laser radiation, especially in the case of a high power, has been the subject of relatively few studies,[14,22,23] in which by no means all possibilities of this method have been considered.

A high intensity of the laser radiation (up to 10^{28} photons/sec in a pulse) and a large cross section for resonance excitation of an atomic transition (10^{-10} cm^2) make it possible in principle to orient nuclei at densities of the nuclei up to 10^{18} cm^{-3}.

Laser radiation with a half-width $\Delta\nu_l$ of the emission line (Fig. 3) consists of modes[24] separated by the distance $\Delta\nu_m$:

$$\Delta\nu_m = c/2L, \tag{3}$$

where L is the length of the laser resonator, and c is the velocity of light. The mode half-width $\Delta\nu_{mw}$ usually varies from a few megahertz to several tens of megahertz. In the cell, the Doppler width $\Delta\nu_D$ of the absorption and emission line profile is about 2 GHz. The natural width of the resonance, corresponding to the lifetime 10^{-8} sec of the excited state, is $\Delta\nu_{lt} \sim 10$ MHz. The field broadening $\Delta\nu_f$ (Ref. 25) at a power of the laser radiation corresponding to the saturation limit of the atomic transition is usually about 10 MHz. The fraction N of atoms in resonance with the laser radiation when $\Delta\nu_m \gg \Delta\nu_D$ is

$$N = \sqrt{\Delta\nu_f^2 + \Delta\nu_{lt} + \Delta\nu_{mw}^2}/\Delta\nu_m. \tag{4}$$

Typical values of the widths are $\Delta\nu_m \sim 150$ MHz, $\Delta\nu_f \gtrsim 10$ MHz, $\Delta\nu_{lt} \sim 10$ MHz, and $\Delta\nu_{mw} \sim 30$ MHz. Substituting these values in (4), we find that $N \sim 24\%$. Since the position of the modes varies with time, the limit for N may be increased. Thus, using laser radiation for optical pumping under the condition $\Delta\nu_l \gg \Delta\nu_D$, without adopting special measures,[26] one can obtain a degree of orientation of about 24%.

The disorientation time is largely determined by the interaction of the excited atoms with one another, with extran-

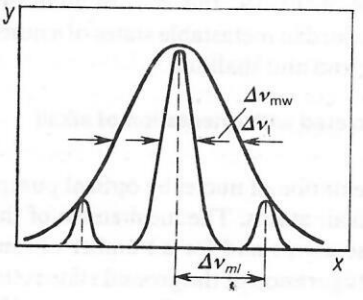

FIG. 3. Structure of radiation generated by a multimode laser (the x axis is the frequency of the laser radiation, and the y axis is the intensity of the radiation).

TABLE I. Table of some characteristic values of the relaxation parameters for polarized atoms.

Relaxation mechanism	Potential	Phenomenon	Cross section cm^2
Dipole–dipole interactions	$(D_A D_B - 3D_A R_{AB} R_{AB} D_B) R^{-3}$	Broadening of optical lines, depolarization in the excited state	$10^{-13} - 10^{-15}$
Spin exchange between electrons	$V(R_{AB}) \mathbf{J}_A \mathbf{J}_B$	Approach of the spin temperature to equilibrium preserves the total spin	10^{-14}
Spin–orbit interaction	$V(R) L \mathbf{J}$	Disorientation of atoms in the S state in collisions with the wall or buffer gas	$10^{-19} - 10^{-26}$
Spin–nucleus interaction	$\mathbf{J} F(R) I$	Disorientation of atoms in the S state in collisions with the wall and buffer gas, polarization of nuclei in spin-exchange collisions	10^{-24}
Nuclear quadrupole interaction	$-\frac{1}{6} \nabla E Q_0$	Relaxation of the spins of diamagnetic atoms in collisions with a wall	Depends on the gradient of the electric field near the wall, the time spent on the wall, and the quadrupole moment of the nucleus

Notation: $\mathbf{J}$ is the electron spin, R_{AB} is a vector connecting two atoms, D_i is the dipole moment of the ith atom, I is the spin of a nucleus, $F(R)$ and $V(R_{AB})$ are the radial dependences of an interaction potential, ∇E is the gradient of the electric field, L is the orbital angular momentum.

eous atoms, and by collisions with the walls of the cell. It is fairly long in the case of atomic beams or vapor for atoms having the S configuration of the electron shell, and also in an atmosphere of inert gases, and is between 10^{-2} sec and several seconds.[9] The characteristic values of the disorientation cross sections are given in Table I. In the case of solids, the disorientation time is, as a rule, very short (10^{-10} sec), and this leads to great difficulties in creating solid targets of oriented nuclei by means of laser radiation.

The process of nuclear orientation can be studied by measuring the anisotropy and polarization of the nuclear radiation or by ensuring the fulfillment of a number of experimental conditions and measuring the angular distribution and polarization of optical radiation. Methods of analyzing the process of optical pumping by means of optical radiation are considered in Refs. 9 and 28.

For the orientation of atoms and nuclei continuous light sources have usually been employed. New possibilities in this direction were opened up in experiments with pulsed dye lasers. For example, their use in Ref. 16 made it possible to demonstrate experimentally that already in one cycle of absorption of polarized photons and spontaneous decay to the ground state one can achieve an appreciable degree of nuclear polarization (10–20% in the case of ^{23}Na), with orientation time $\tau_{or} \sim 3 \times 10^{-8}$ sec. The influence of various external conditions on the degree of orientation of the nuclei was also studied in Ref. 16. The degree of orientation of the ^{23}Na nuclei is shown as a function of the power of the laser radiation in Fig. 4. The greatest degree of orientation was observed when the power corresponded to the saturation limit of the atomic transition. When the power was increased, the degree of orientation of the nuclei was observed to decrease. This decrease can be explained as follows. Since the duration of the laser pulse was 10^{-8} sec, and the lifetime of the excited atomic level ($P_{1/2}$) is $\sim 1.6 \times 10^{-8}$ sec, in the optimal case the maximal number of Na atoms is in the excited state at the moment when the laser pulse ends, and the atoms then decay spontaneously.

To eliminate the influence of extraneous magnetic fields on the orientation of the atom and the nucleus, one can either use compensation of the fields or apply a stronger magnetic field in the required direction, making the atoms begin to precess around the total magnetic field without losing their orientation. Thus, to eliminate the influence of weak (up to 1 Oe) magnetic fields, a stronger magnetic field (up to 100 Oe) directed along the propagation axis of the laser radiation was applied in Ref. 14. The degree of orientation $P_I = \langle I_z \rangle / I$ of the nuclei was measured as a function of the applied external magnetic field. For zero value of the applied field, an orientation signal was not observed: $P_I = 0$ (Fig. 5). This is explained by the fact that the atoms precess around the Earth's magnetic field, which was directed perpendicular to the propagation axis of the laser radiation. As the external magnetic field was increased in strength, a growth in the degree of orientation P_I was observed.

Orientation of long-lived isotopes in a cell

When a dye laser is used to obtain and study oriented nuclei of radioactive elements, one must distinguish long- and short-lived nuclei. For the former, the cell walls and relaxation processes on them begin to play an important part. The orientation of the long-lived ^{22}Na and ^{24}Na nuclei in a cell by means of pulsed laser radiation was studied in Refs. 14 and 16. The problem reduced to the following:

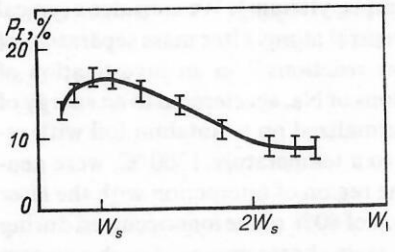

FIG. 4. Degree of orientation of Na nuclei as a function of the power of the laser radiation (W_1 is the power of the laser radiation, and W_s is the power of the laser at the saturation limit of the atomic transitions).

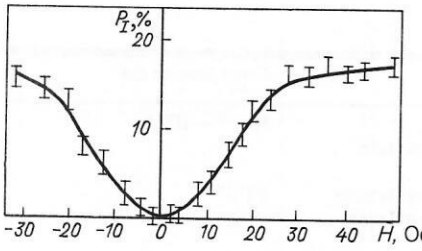

FIG. 5. Degree of orientation P of the nuclei as a function of the external magnetic field H.

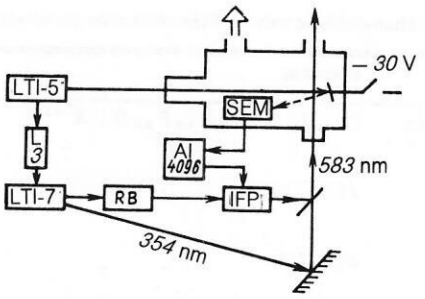

FIG. 6. Experimental arrangement intended for investigation of the process of pulsed evaporation and decomposition of substances.

1. One must obtain vapor of Na neutral atoms with only a slight admixture of Na isotopes in the form of different chemical compounds, since the latter do not participate in the process of optical pumping.

2. The cell must not be heated above 250 °C in order to minimize the effects of disorientation on the walls.

3. One must use materials with a low disorientation cross section, transparent for the optical radiation, and capable of ensuring a sufficiently high vacuum in the cell (10^{-4} Pa).

4. It is necessary to create conditions in the experimental apparatus that lead to a minimal contribution of depolarization effects. In addition, the number of atoms used for orientation must not exceed $\rho_{eq} V$, where ρ_{eq} is the equilibrium density of the atoms at the given wall temperature of the cell, and V is the volume of the cell. If this is not done, an appreciable fraction of the atoms will be on the walls and will give a background. The unsuccessful attempts to obtain oriented nuclei of long-lived nuclides in a number of studies[17] are due, in our opinion, to the attempt to orient a large number of atoms.

At cell wall temperature $t = 200$ °C and volume $V = 1$ cm^3, the number of atoms must be limited (in the case of Na), $N \lesssim 10^{13}$, this value corresponding to an activity of about 5 μCi for ^{22}Na. In Refs. 14 and 16, the difficulties of preparing samples of the investigated substances were overcome by means of pulsed laser evaporation and decomposition of various chemical compounds in vacuum. We shall consider this question in somewhat more detail.

Laser radiation is currently used to investigate radioactive nuclei in neutral atoms or ions.[7] In the majority of cases, neutral atoms are required for experiments. Usually, the ions of the investigated substances, obtained as a result of nuclear reactions, are transported to the region of interaction with the laser radiation, prior to which they are decelerated in various foils, and then, the final foil being heated, the substances evaporate in the form of neutral atoms.[6] If one is working with elements having a low ionization potential, the surface of the foil must be covered by a material with a low work function (for example, yttrium). We consider a typical scheme for obtaining neutral atoms after mass separation of the products of nuclear reactions[30] in an investigation of short-lived Na nuclei. Ions of Na, accelerated to an energy of about 60 keV, were thermalized on a tantalum foil with yttrium covering heated to a temperature 1200 °C, were neutralized, and entered the region of interaction with the laser radiation. Neutralization of 80% of the ions occurred during a time ~150 msec. The main shortcomings of such a system for obtaining oriented nuclei of long-lived Na isotopes are the need to heat the foil to high temperatures and to use materials with a large disorientation cross section, a fact that requires one to work with only atomic beams.

However, it is known that if the sample is irradiated with the irradiation of the first harmonic ($\lambda = 1060$ nm) of a Q-switched YAG:Nd$^+$ laser, evaporation of the matter in the form of neutral atoms and ions occurs.[31] It is noted in Ref. 32 that when salts are decomposed in this way up to 70% of the evaporated matter may be in the form of neutral atoms and the remaining 30% in the form of ions, the evaporation process lasting for several hundred nanoseconds. Such an evaporation regime, used with the aim of obtaining neutral atoms for subsequent investigation by means of laser radiation, was studied experimentally in Ref. 33 by means of the arrangement shown in Fig. 6.

The investigated substance (NaCl) was deposited on a zinc plate. Infrared radiation of the YAG:Nd$^+$ laser was focused by means of a lens onto a sample in a spot with diameter 1 mm. The energy density of the incident radiation was 0.1 J/cm^2. The duration of the laser pulse was 10^{-8} sec. A dye laser was pumped by the second harmonic ($\lambda = 532$ nm) of another YAG:Nd$^+$ laser, whose third harmonic ($\lambda = 353$ nm) served to ionize the excited neutral atoms obtained as a result of the laser evaporation and decomposition of the investigated substance. The dye laser was tuned to the frequency of the $S_{1/2} \rightarrow P_{1/2}$ transition in sodium. The width of the laser emission line was $\Delta\nu \sim 20$ GHz. Two Fabri–Pero interferometers were used, one of which was placed within the dye laser and reduced the width of the emission line to 1 GHz, while the other, placed outside the resonator in a pressure chamber, reduced the width of the emission line to 200 MHz and made it possible to adjust the wavelength of the laser radiation in a range of 6 GHz. The ions formed as a result of the resonance absorption of the laser radiation and subsequent ionization (Figs. 6 and 7) were detected by a

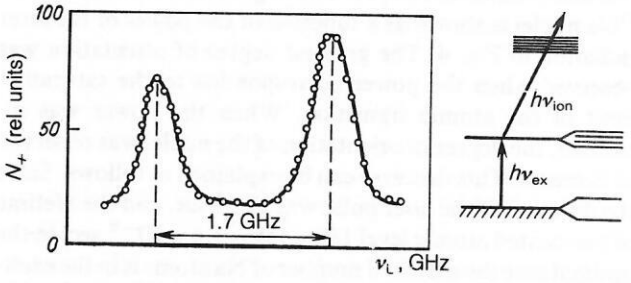

FIG. 7. Spectrum of hyperfine splitting of the D_2 line of ^{23}Na obtained by means of a laser atomizer[67]; N_+ is the number of counts of the secondary electron multiplier as the frequency ν_1 is scanned.

secondary electron multiplier (VÉU-6). The dependence of the ion current on the frequency of the laser radiation is determined by the structure of the levels of the ^{23}Na atoms. Figure 7 gives the spectrum of the hyperfine splitting in the D_2 line of ^{23}Na obtained by such a method. The experimental signal-to-noise ratio was 3:1, indicating that about 67% of the evaporated matter was in the atomic state. When a blocking potential -30 V was applied to the substrate with the sample, the signal-to-noise ratio was improved to 10:1, indicating that 33% of the evaporated substance was produced in the form of ions.

Processes of orientation relaxation in the cell

We consider a number of questions relating to the relaxation of the orientation in the cell. At low pressures of the residual gas, when the mean free path of the atoms is greater than the cell diameter, the main contribution to the relaxation is made by processes on the walls. However, it is an important fact that for atoms oriented in the cells there may be many hundreds of collisions with molecules of the buffer gases or with a wall without loss of orientation.[34] This is true only for atoms for which the electron shell is in the S state, since the spherical distribution of the charge in the atom protects the spins from disorientation effects in collisions.

The spin-disorientation cross section of nuclei colliding with the cell walls is 10^{-19}–10^{-26} cm^2 and depends on the wall material.[9] For example, for glass and quartz the number of collisions without loss of orientation is 10^2–10^3, for a paraffin covering 10^3–10^4, and for special coverings 10^4–10^5 (Refs. 8, 9, 13, and 34; see also Table I). In a number of cases, spin disorientation occurs after a much larger number of collisions. The spin-orientation relaxation of cesium and rubidium atoms requires 10^6–10^9 collisions of the atoms with one another. It was shown in Ref. 36 for the first time that the atoms of alkali metals can be oriented by means of optical radiation in an evacuated volume if the glass walls of the chamber are covered by materials with a small disorientation cross section. The relaxation time was of the order of a second. It was found that paraffins and various organic compounds are particularly effective in the case of optical pumping of atoms of alkali metals. It was also shown that the relaxation rate is lower for paraffin coverings containing deuterium. This fact indicates that the magnetic fields of the nuclei on the cell walls play an appreciable part in the relaxation processes, since the magnetic moment of the deuteron is approximately three times smaller than the magnetic moment of the proton.

However, the complete process of the spin-orientation relaxation of the atoms cannot be exclusively attributed to the magnetic fields of the atoms and nuclei on the walls of the cell. Other processes can also play an important part. Thus, it was shown in Ref. 37 that the polarization of the hyperfine-structure components $\langle \mathbf{J}\cdot\mathbf{I}\rangle$ relaxes at the same rate for both ^{85}Rb ($\mu = 1.353$) and ^{87}Rb ($\mu = 2.751$) on deuterium coverings but is slightly different for hydrogen coverings. This is explained by the fact that for the deuterium coverings, where the relaxation rate in the magnetic fields of the walls is small, the main contribution to the relaxation of $\langle \mathbf{J}\cdot\mathbf{I}\rangle$ is made by the spin–orbit interaction with the atoms of the walls. For walls with hydrogen covering, a significant contribution to the relaxation rate is made by interaction with the magnetic fields of the cell walls, and, since the magnetic moments of the ^{85}Rb and ^{87}Rb nuclei are different, a difference is observed in the relaxation rates. The disorientation cross section depends strongly on the wall temperature, and when the latter is increased it increases too as a rule.[38] We note that data are often given in the literature on the spin-disorientation cross sections of an electron shell or an atom but not a nucleus. As a rule, the nuclear spin relaxes much more slowly than the spin of the electron shell.

The mechanism of the disorientation process depends strongly on the ratio of the times of interaction with the depolarizing fields in the collisions and on the following characteristic times:

(1) the hyperfine-interaction time $\tau_{\text{hpf}} \sim 10^{-10}$ sec;

(2) the fine (spin–orbit) interaction time $\tau_f \sim 10^{-13}$ sec;

(3) the collision time $\tau_c \sim 10^{-12}$ sec.[9]

Since $\tau_{\text{hpf}} \gg \tau_c$ and $\tau_T \ll \tau_c$, the hyperfine interaction does not feel such a rapid perturbation, whereas the fine interaction can carry the system to equilibrium. Therefore, in collisions of duration less than τ_{hpf} the spin of the nucleus does not change its direction appreciably, whereas the spin of the electron shell can be completely disoriented. The nuclear spin inertia in collisions of atoms of alkali metals was established experimentally in Ref. 39. The experimental value of the spin-disorientation cross section of nuclei in collisions with the walls (quartz and special coverings) is $\sigma \sim 10^{-22}$–10^{-26} cm^2.[9] The cross section of polarization of nuclei of alkali metals in spin-exchange collisions of the atoms with one another is of the same order of magnitude: $\sigma \sim 10^{-24}$ cm^2. The main contribution to the depolarization of the electron shell is made by spin-exchange collisions with extraneous impurities ($\sigma \sim 10^{-14}$ cm^2), for example, with the residual air molecules. As a result of spin-exchange collisions, the spin orientation of the electron shell does not, in general, disappear, but is passed from atom to atom. The disorientation cross section of the electron-shell spins in collisions of atoms of alkali metals with one another is $\sigma \sim 10^{-17}$ cm^2.[40] If the cell is evacuated to pressures 10^{-4} Pa and purified of chemically active elements, these depolarization factors can be almost completely eliminated. In this case, the main contribution to the spin relaxation of the nuclei is made by the processes on the cell walls. The relaxation is determined by the time during which the atom is on the cell walls, by the local magnetic field, by the gradient of the electric fields, and by the nuclear quadrupole and magnetic moments. The mean time that an atom is on a cell wall is[9]

$$\tau_w = \tau_0 \exp(E_a/kT), \tag{5}$$

where $E_a \sim 0.1$ eV is the energy of stripping of the atom from the wall (for quartz), T is the wall temperature, and $\tau_0 \sim 10^{-12}$ sec is the period of the vibrations of the atoms about the equilibriun position in the case of interaction with the wall.

For $T = 473$ °K and $\tau_w = 10^{-11}$ sec, i.e., less than the hyperfine interaction time ($\tau_{\text{hpf}} \sim 10^{-10}$ sec), this, in its turn, means that it is mainly the electron shell that is disoriented in the collisions with the walls.

To investigate the process of orientation of Na nuclei by optical pumping in a cell the optimal conditions correspond to heating of the walls to $t \sim 150$–200 °C. Since with decreasing temperature the time during which an atom is on a wall

increases, and with it the disorientation probability, but the chemical activity of the walls increases with the temperature, and this also destroys the orientation, it is necessary to select the optimal temperature regime.

In Ref. 14, the mean flight time of the Na atoms from wall to wall was $\tau_{fl} \sim 3 \times 10^{-5}$ sec, i.e., much greater than the time during which an atom remains on a wall. Therefore, all the foregoing arguments are correct only when the number N of atoms in the cell satisfies the condition

$$N \leqslant \rho_{eq} V, \tag{6}$$

where ρ_{eq} is the density of the saturated Na vapor at the given wall temperature, and V is the cell volume. If this condition is not satisfied, the majority of atoms will be on the cell walls and will not participate in the orientation process. Therefore, it is desirable to work with sources with a minimal amount of stable isotopes of the investigated elements.

Investigations with optically oriented isotopes

To obtain oriented Na nuclei, an optically transparent layer of Na vapor was excited by circularly polarized light with wavelength $\lambda = 589.6$ nm (D_1 transition). The width of the irradiation line was 20 GHz, which covered the isotopic shifts of ^{22}Na($-$ 757 MHz), ^{24}Na(706 MHz), and ^{23}Na, the hyperfine splitting, and the Doppler broadening and also ensured that the D_2 transition was avoided (480 GHz). The power of the laser radiation was chosen to ensure the regime of saturation of the atomic transition, i.e., after the time of action of the laser pulse all the atoms in the region of illumination had been resonantly excited. In the subsequent decay, about 8% of ^{22}Na atoms and 6% of the ^{24}Na atoms went over to the $S_{1/2}$ level with maximal projection of the angular momentum.

In work with the radioactive isotopes, the angular distribution of the γ rays from the decay of the oriented nuclei was measured. In the case of the stable isotopes, the fluorescence signals, detected by two photomultipliers, in front of which rotating polaroids and $\lambda/4$ plates were placed, were studied. For the measurement of the γ anisotropy, four γ spectra were measured for the same detector position and for the same time interval (Fig. 8):

1. The γ spectrum in the presence of the laser beam and a magnetic field applied along the direction of propagation of the laser beam: $N(\theta, \sigma^+, H^+)$.

2. The γ spectrum without the laser beam (covered by a diaphragm D) but with the same magnetic field: $N(\theta, 0, H^+)$.

3. The γ spectrum in the presence of the laser beam and with a magnetic field in the opposite direction to the beam: $N(\theta, \sigma^-, H^-)$.

4. The γ spectrum without the laser beam: $N(\theta, 0, H^-)$.

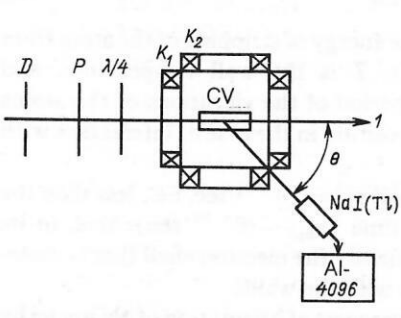

FIG. 8. Arrangement of experiment intended to measure the angular distribution of the γ radiation of nuclei oriented by means of a dye laser: 1 is the laser beam, D is a diaphragm, P is a polaroid, $\lambda/4$ is a quarter-wave plate, K is a Helmholtz coil, and CV is the cell with Na vapor.

Such a method makes it possible to eliminate the corrections for the change in the solid angle and γ detection efficiency, and also the corrections to take into account the influence of the magnetic field on the photomultiplier. Some control experiments in which no orientation of the ^{22}Na, ^{24}Na, and also the ^{23}Na nuclei should be observed were made. These involve measurement of the γ angular distribution at frequencies of the laser radiation different from the resonance frequency, measurement of the angular distribution of the γ rays and the fluorescence in uncompensated magnetic fields, and measurement of the γ angular distribution without the pulsed decomposition of the Na salts.

The ratios

$$W^{\pm}(\theta) = \frac{N(\theta, \sigma\pm, H\pm)}{N(\theta, 0, H\pm)} \tag{7}$$

give the γ angular distribution in pure form. Degrees of orientation $\sim 12\%$ for ^{22}Na and $\sim 8\%$ for ^{24}Na were obtained.

Effects associated with parity nonconservation in weak interactions are manifested in annihilation radiation following β^+ decay of oriented nuclei.[41] If unoriented nuclei decay, the positrons are emitted isotropically, and the ratio (7) for the annihilation radiation must be equal to unity for all angles θ. This indicates that the experiment has been correctly performed. This was confirmed by measurements in zero magnetic field.

Because parity is not conserved in β^+ decay of oriented nuclei, an asymmetry of the emission of the β^+ particles is observed. The angular distribution of the β^+ particles in the model with $V-A$ interaction has the form

$$W(\theta) = 1 + P_I a \frac{\langle v \rangle}{c} \cos\theta, \tag{8}$$

where $a = 1$ for β^+ decay with $\Delta I = 1$, $\langle v \rangle$ is the mean velocity of the β^+particles ($\langle v \rangle/c \approx 0.6$ for ^{22}Na), and P_I is the degree of orientation of the nuclei. Since the solid angles for detection of the annihilation radiation are different for different points at which the annihilation occurs, the asymmetry of the β^+ radiation can be reflected in an asymmetry of the annihilation radiation. For the cell used in Ref. 14, the geometrical conditions were the same for the angles 0° and 90°. Therefore, exact measurement of the asymmetry is made possible by comparing the effects at these angles. The experimental value of the asymmetry $A_\beta = P_I \langle v \rangle/c$ was found to be $A_\beta^+ = 0.05 \pm 0.01$. The sign of the effect was reversed when the direction of the spin orientation of the nuclei was reversed,

$$A_{\bar{\beta}} = -0.04 \pm 0.01, \tag{A}$$

whereas the anisotropy of the γ rays with energy 1280 keV did not change sign (Fig. 9). This fact confirms the conclusion that the asymmetry observed in the annihilation radiation is due to parity nonconservation in the β^+ decay. The theoretical calculation of the angular distribution of the annihilation radiation for angles different from 0° and 90° is made difficult by the fact that the geometrical factors are different and, in addition, it is difficult to calculate the quan-

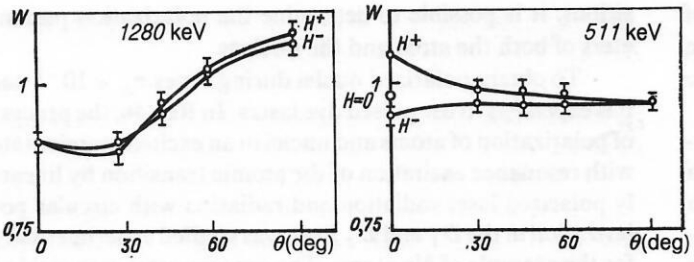

FIG. 9. Angular distribution of γ rays from the decay of ^{22}Na nuclei oriented by means of a dye laser.

tities associated with the reflection of the β^+ particles by the cell walls. Therefore, in the experiment the angular dependence for the annihilation radiation agreed with (8) only in sign. In Ref. 14, the number of collisions with the cell walls during which disorientation of the nucleus did not occur was determined and found to be $N \gtrsim 3 \times 10^3$, and the cross section of disorientation on the wall was $\sigma \lesssim 10^{-24}$ cm^2.

In Ref. 20 there is a description of the method of obtaining oriented nuclei of the ^{240m}Am shape isomer by means of laser radiation. The asymmetry of the fission fragments of the ^{240m}Am nuclei was measured as a function of the frequency of the laser radiation. When the Am atoms were excited by polarized laser radiation, they were oriented, and this was reflected in the angular distribution of the fission fragments. The ^{240m}Am nuclei were obtained in the ^{238}U(^{7}Li, $5n$) reaction. The $^8S_{7/2} \rightarrow {}^{10}P_{7/2}$ atomic transition was excited and the optical isomer shift was measured. From the results of the measurements, Benis *et al.*[20] succeeded in estimating the change in the mean-square radius of the ^{240m}Am and ^{240}Am nuclei, $\delta\langle r^2\rangle \sim 5$ F^2, a result that confirms the conclusions drawn from calculations by Strutinskiĭ's method[42] about the double-hump nature of the fission barrier in the Am nucleus. For further comparison with theory, it would be desirable to measure the hyperfine structure in ^{240m}Am atoms and to determine the quadrupole moment of the shape isomer.

The atomic transition in Am was excited by laser radiation from a cw dye laser operating in the multimode regime with line half-width 0.05 nm and density 5 W/cm^2. In the atomic ground state $^8S_{7/2}$, the hyperfine splitting is small, and this means that to obtain oriented Am nuclei one cannot allow disorientation in the atomic excited state $P_{7/2}$. However, the $P_{7/2}$ state does not have spherical symmetry, and this leads to a sharp growth of the disorientation cross sections in collisions,[9] which, in its turn, imposes rather stringent conditions on the admissible concentrations of impurities in the region of observation and on the use of buffer gases. Unfortunately, Ref. 20 is as yet the only publication devoted to orientation of Am nuclei by optical pumping.

Thus, by means of laser radiation it is possible to orient both long- and short-lived nuclei of different elements without using the technique of low temperatures and strong magnetic fields. One of the important advantages of this method is that it enables one to obtain an appreciable degree of orientation of the nuclei during a time of 10^{-8} sec. The shortcomings of the method include the restrictions associated with the need to use volatile substances with the S configuration of the electron shell. In addition, significant numbers of nuclei cannot be oriented in cells.

The use of the method of nuclear orientation by optical pumping appears to be most promising in the case when one wishes solve physical problems that require selectivity with respect to the oriented elements and a high speed.

POLARIZATION OF NUCLEI BY EXCITATION OF AN ATOMIC TRANSITION WITH POLARIZED LASER RADIATION

In the process of optical pumping of atoms by polarized light the spins of the atoms and nuclei are polarized in the ground or a metastable atomic state. The time during which the nuclear spin is oriented (τ_{or}) in this method is limited by three quantities:

(1) the stimulated-transition time (τ_{st})

$$\tau_{or} > \tau_{st}; \tag{9}$$

(2) the time of spontaneous decay from the excited atomic state:

$$\tau_{or} \gtrsim \tau_{st} + \tau_{sp}; \tag{10}$$

(3) the maximal hyperfine splitting ΔE_{hpf} in the ground or excited atomic states and the associated hyperfine-interaction time ($\tau_{hpf} \sim h/\Delta E_{hpf}$):

$$\tau_{or} > \tau_{hpf}. \tag{11}$$

For a fairly large number of atoms it is possible to choose optical transitions such that $\tau_{hpf} < \tau_{or}$,[43,44] and by exciting the atomic level by laser radiation one can fairly easily satisfy the relation $\tau_{st} \ll \tau_{sp}$.[45] In this case, the time during which the orientation of the nuclei in the atomic ground state takes place will be bounded below only by the spontaneous-decay time: $\tau_{or} > \tau_{sp}$. The typical value of τ_{sp} for allowed $E1$ transitions in an atom is about 10^{-8} sec, and this, in its turn, means that the optical pumping method cannot be used to orient short-lived nuclear states (with lifetime less than 10^{-8} sec). However, since we can transfer an atom from the ground to an excited state in a time much shorter than 10^{-8} sec, and can also regulate this time (τ_{st}) by choosing an appropriate power of the laser radiation, it is interesting to consider the process of nuclear polarization in an excited atomic state.

The process of orientation of nuclei in an excited atomic state during a time $\tau_{or} < \tau_{sp}$ was analyzed theoretically for the first time in Ref. 22 for the case when there is a hyperfine interaction in the ground atomic state but the hyperfine splitting in the excited atomic state can be ignored. It was shown that to obtain the maximal degree of orientation it is necessary to excite a $J \rightarrow J$ transition (D_1 transition in alkali atoms); the degree of orientation of the nuclei in this case is roughly doubled ($J = 1/2$) compared with excitation of $J \rightarrow J+1$ transitions (D_2 line), and the maximal degree of

orientation is 6%. The time during which the orientation of the nucleus occurred was equal to the stimulated-transition time and was limited by the hyperfine splitting in the atomic ground state.

For orientation of nuclei in either the ground or an excited state it is essential to have in at least one of these states a hyperfine interaction between the nucleus and the electron shell, by means of which the angular momentum of the photon absorbed by the atoms is distributed between the nucleus and the electron shell. Polarization of nuclei following excitation of an atom by polarized laser radiation can occur only in the following three cases:

(1) there is hyperfine structure in the excited atomic state;

(2) there is hyperfine structure in the ground atomic state;

(3) there is hyperfine structure in both the ground and the excited atomic states.

In Ref. 46, polarization of nuclei in an excited atomic state in the presence of hyperfine splitting in both states (case 3) was considered. In this situation, one can achieve a higher degree of nuclear polarization than in cases 2 and 1, although the orientation time (τ_{or}) is bounded below by two quantities—the hyperfine-interaction times in the ground and excited atomic states (τ_{hpf}^{g}, τ_{hpf}^{ex}). The polarization in the excited atomic state is due both to the polarization of the exciting light and to the polarization in the ground atomic state.

In Ref. 46 experiments are described in which the ground state at the initial time was not polarized and the excitation was done by means of a broad line, i.e., the halfwidth of the laser emission line ($\Delta\nu_l$) exceeded the hyperfine splitting in both the ground and the excited atomic states. In this case, orientation and alignment occur in the electron shell. If the hyperfine-interaction time is less than the lifetime of the atom in the excited atomic state, then the electron polarization is transferred to the nucleus.

When the atom is excited by a broad line, the appearance of the nuclear polarization can be interpreted as a process of relaxation of the electron-shell polarization with hyperfine interaction superimposed.[47] The angular distribution and the polarization of the fluorescence in optical transitions will depend on whether or not the nucleus was polarized during the lifetime in the excited atomic state. Indeed, the angular distribution and the polarization of the fluorescence depend on the degree of polarization of the electron shell, and in the case of a hyperfine interaction the spin of the electron shell begins to precess around the total spin F, and this, in its turn, signifies the occurrence of polarization of the atom and the nucleus and a decrease of the electron-shell polarization, this being reflected in the angular distribution and polarization of the optical radiation, i.e., the hyperfine interaction depolarizes the fluorescence and smooths the angular distributions. The calculations of Ref. 69 show that the polarization of the fluorescence can be reduced by several times. With increasing polarization of the nucleus and the atom (optical pumping) and the approach to a steady limit, the degree of polarization of the fluorescence also increases, but, as already noted, this process requires a longer time.

Studying the polarization and angular distribution of the fluorescence and knowing the scheme of the optical transitions, it is possible to determine the polarization parameters of both the atom and the nucleus.

To obtain polarized nuclei during times $\tau_{or} < 10^{-8}$ sec it is expedient to use pulsed dye lasers. In Ref. 46, the process of polarization of atoms and nuclei in an excited atomic state with resonance excitation of the atomic transition by linearly polarized laser radiation and radiation with circular polarization in the D_1 and D_2 lines was studied experimentally for the example of Na atoms. The experimental data yielded an estimate of the degree of polarization of the Na nuclei in the excited atomic state, and a comparison was made with the corresponding degree of polarization obtained after decay to the ground state.

We now consider the formation of polarized atoms and nuclei when polarized photons are absorbed, and we analyze the angular distribution and the polarization of the fluorescence for the subsequent radiation on the decay to the ground state.

We shall direct the quantization axis oz and the external magnetic field along the laser beam in the case of excitation by circularly polarized radiation and along the direction of the polarization vector of the light in the case of linear polarization of the laser radiation. Then, in accordance with the selection rules for the magnetic quantum number, a transition from some sublevel $|FM\rangle$ of the ground state of the atom, following absorption of one photon, is possible to not more than one sublevel $|F^*M^*\rangle$ of the excited state, i.e., the conditions of the experiment are chosen in such a way that coherent polarization effects, which will be discussed in the following section, are absent and the density matrix of the excited state of the atom is diagonal with respect to the magnetic quantum numbers:

$$\rho_{MM'} = \delta_{MM'}\rho_M. \qquad (12)$$

This means that the phases of atomic states that differ in the magnetic quantum number are uncorrelated, and for such a geometry of the experiment we study effects associated with the difference of the populations over the different Zeeman sublevels in the excited atomic state.

The schematic distribution of the populations of the magnetic sublevels of the Na atoms in the excited atomic state in the absence of depolarization effects is shown in Fig. 10. This reflects the case when the excitation is done by a broad line. Then, after absorption of linearly polarized light in the D_1 line, the angular distribution of the fluorescence is isotropic and there is no polarization.[48] This fact is used in control experiments.

In the case of irradiation with circularly polarized light in either a D_1 or a D_2 transition an orientation of the electron shell and the nucleus occurs already after absorption of a single photon in the atom. In the case of irradiation with linearly polarized light in a D_2 transition, alignment occurs in the system. One must also take into account the contribution to the polarization from the ground atomic state. For example, in the case of irradiation with σ^+ light in the D_1 line the $E1$ transitions from the level $F = 2$, $m_F = 2$ are forbidden by the selection rules, whereas they are allowed from the remaining Zeeman sublevels. At the same time, it is necessary to take into account the contribution of the state $F = 2$, $m_F = 2$, $S_{1/2}$ to the total polarization.

To get a clearer picture of the polarization mechanism, the Rabi oscillations are often ignored.[49] In fact, the oscilla-

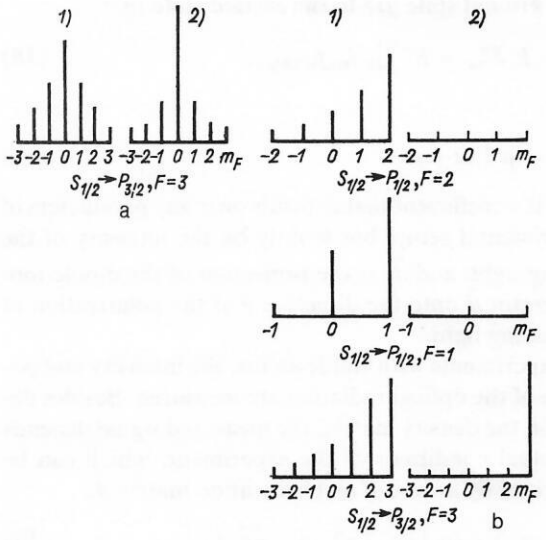

FIG. 10. Population of magnetic sublevels of ^{23}Na atoms in an excited state following illumination with polarized light: (a) with linearly polarized light, (b) with circularly polarized light; (1) the step; (2) the steady limit.

tions in the populations of the magnetic sublevels take place over a time $(10–30)\tau_{sp}$, and to obtain extremal values of the polarization it is necessary to choose the power of the laser radiation appropriately.

The experiments reported in Ref. 46 were done in the optimal regime, i.e., during the time of action of the laser pulse approximately one stimulated transition occurred in each atom. Then at the time when the laser pulse ends the greatest number of atoms is in the excited state. The following values were obtained in the experiments: A_2^I to 0.14% and P_I to 0.11% in the D_2 transition, and P_I to 0.31% in the D_1 transition; here

$$\left.\begin{aligned} A_2^I &= \sum_{FM} \rho_{FM} \langle FM|3I_z^2 - I^2|FM\rangle / I(2I+1), \\ P_I &= \sum_{FM} \rho_{FM} \langle FM|I_z|FM\rangle / I, \end{aligned}\right\} \quad (13)$$

where ρ_{FM} is a diagonal element of the density matrix of the atom. The orientation time was about 10^{-8} sec.

COHERENT EFFECTS IN THE ATOMIC SHELL AND THE PHENOMENON OF LEVEL CROSSING FOR ATOMS WITH RADIOACTIVE NUCLEI

In this and the following section, we consider questions related to polarization coherence, namely, we analyze processes of obtaining ensembles of atoms and nuclei for which the states differing in the magnetic quantum number are connected in phase, and we investigate the decay of such systems. Our main attention will be devoted to the phenomenon of level crossing in the electron–nucleus system and its manifestation in the optical and nuclear radiations.

It should be noted that these questions illuminate only a small number of the methods of coherent optics that could be used in nuclear physics.[23] These methods include, in the first place, methods associated with spatial coherence: photon echo, double resonance, superradiance, etc. Development of these methods at the present time, in which pulsed and cw dye lasers are available, promises a significant extension of the ideas and methods of optics in nuclear physics.

Polarization coherence in the atomic shell and interference phenomena in optical radiation

We consider a number of phenomena that arise in optical radiation because of the existence of a phase connection between atomic sublevels, i.e., coherence, and we analyze the possibilities of some methods associated with coherence in the atomic shell for the determination of nuclear characteristics.

For $E1$ transitions in an atom the intrinsic polarizations are $\sigma^+(\Delta m = 1)$, $\sigma^-(\Delta m = -1)$, and $\pi(\Delta m = 0)$, i.e., e_{+1}, e_{-1}, e_0 are the orthogonal components of an irreducible tensor of third rank. The polarization of laser radiation can be expanded with respect to these components:

$$e = a_+ e_{+1} + a_{-1} e_{-1} + a_0 e_0. \quad (14)$$

If in the expansion (14) only one coefficient is nonzero, then on the absorption of a photon from a given magnetic sublevel of the atomic ground state only one magnetic sublevel of the excited state can be populated. Such polarization of the light is said to be incoherent. If in the expansion (14) at least two coefficients are nonzero, then two sublevels of the excited state can be populated, and the phases of the populated states are correlated. Such polarization of light is called coherent. The presence of states with correlated phases leads to a number of interference phenomena in the spontaneous emission of atoms and is manifested in changes of the polarization, angular, and time characteristics of the optical radiation. We have here in the first place phenomena such as[53] level crossing, quantum beats, paramagnetic resonance, galvano-optics,[54] and others.

We give examples of light with coherent and incoherent polarization. As the quantization axis, we take the direction oz of the external magnetic field. In the case of linearly polarized light [Fig. 11(a)] and a magnetic field directed along the polarization vector e_0, only the single coefficient a_0 in the expansion (14) is nonzero, but if the magnetic field is directed along the laser beam, then in the expansion (14) there will already be two nonvanishing coefficients, a_+ and a_- (since linearly polarized light can be regarded as the sum of two circular polarizations), and it is possible to have pop-

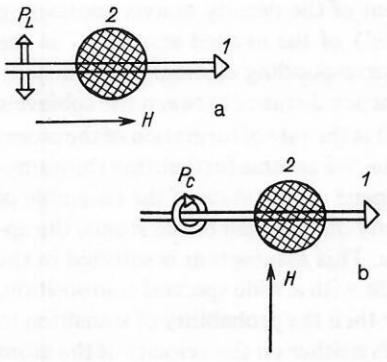

FIG. 11. Arrangement for obtaining ensembles of atoms and nuclei with polarization coherence: (a) in the case of illumination of atoms with linearly polarized light P_L; (b) in the case of illumination of atoms with circularly polarized light P_c; 1 is the laser beam, 2 is the region of interaction with the atoms, and H is the direction of the magnetic field.

ulation of two sublevels (i.e., the formation of a superposition state) in the excited atomic state differing in their magnetic quantum number by 2 ($\Delta m = 2$ is the coherence).

In the case of excitation by circularly polarized light σ^+ [Fig. 11(b)] and a magnetic field directed along the laser beam, only the coefficient a_+ is nonzero in the expansion (14), and we cannot excite a coherent superposition of states in the atom. But if the magnetic field is directed perpendicular to the laser beam, three coefficients a_+, a_0, a_- will be nonzero, and we can excite a coherent superposition of states with $\Delta m = 1, 2, 3$.

We now give a brief description of interference phenomena in the optical radiation. For stable atoms, a detailed analysis of these phenomena is given in Ref. 53. We shall give the description in the formalism of the density matrix ρ. To simplify the mathematical description, we consider the most elementary conditions for performing the physical experiment. We assume that the atom can be in two states, the ground state with sublevels μ and the excited state with sublevels n. We restrict ourselves to conditions under which optical coherence, i.e., coherence between the sublevels of the ground and excited states, does not arise; this means vanishing of the matrix elements

$$\rho_{\mu n} = 0. \tag{15}$$

We give the treatment in the first order of perturbation theory. This condition is satisfied in the case of excitation by a weak optical field with a wide spectrum. We also assume that the number of atoms in the excited state is small compared with the number of atoms in the ground state. In this case, the density matrix can be decomposed into two parts: the density matrix of the ground state and the density matrix of the excited state. Under such conditions, the density matrix of the ground state can be regarded as constant, independent of the applied optical field. In addition, in all that follows, unless specially stated otherwise, we shall assume that the density matrix of the ground state does not contain nondiagonal terms, i.e., the ground state does not possess coherence:

$$\rho_{\mu\mu'} = 0, \quad \mu \neq \mu'. \tag{16}$$

The equation for the density matrix in this case has the form[53]

$$\frac{d}{dt}\rho_{nn'} = -\Gamma_{nn'}\rho_{nn'} - i\omega_{nn'}\rho_{nn'} + F_{nn'}(t), \tag{17}$$

where $\rho_{nn'}$ is the element of the density matrix connecting the sublevels $|n\rangle$ and $|n'\rangle$ of the excited state, $\Gamma_{nn'}$ is the decay constant of the corresponding element of the density matrix, $\omega_{nn'}$ is the frequency distance between the sublevels $|n\rangle$ and $|n'\rangle$, and $F_{nn'}(t)$ is the rate of formation of the atoms in the superposition state. We assume further that the pumping $F_{nn'}(t)$ does not depend on the state of the ensemble of atoms, i.e., on the velocity distribution of the atoms, the applied external fields, etc. This assumption is satisfied in the case of excitation by light with a wide spectral composition, i.e., by a broad line. For then the probability of transition to the excited state depends neither on the velocity of the atom nor on the shift of the resonance frequency under the influence of the external fields. Under conditions of irradiation by a broad line, the pumping matrix $F_{nn'}$ can be expressed simply in terms of the matrix element of the $E1$ transition $f_{\mu n}$ from the ground state $|\mu\rangle$ to the excited state $|n\rangle$:

$$F_{nn'} = K'F^0_{nn'} = K'\sum_\mu f_{\mu n}f_{n'\mu}\rho_{\mu\mu}, \tag{18}$$

where

$$f_{\mu n} = \langle \mu \mid \hat{d}\hat{e} \mid n \rangle. \tag{19}$$

Here, K' is a coefficient that depends on many parameters of the experimental setup, but mainly on the intensity of the irradiating light, and $\hat{d}\hat{e}$ is the projection of the dipole-moment operator $\hat{d}$ onto the direction $\hat{e}$ of the polarization of the irradiating light.

In experiments with stable atoms, the intensity and polarization of the optical radiation are measured. Besides depending on the density matrix, the measured signal depends on the actual conditions of the experiment, which can be characterized by means of an observation matrix $\hat{A}$:

$$A_{nn'} = \sum_\mu \langle \mu \mid \hat{d}\hat{e}_0 \mid n \rangle \langle n' \mid \hat{d}\hat{e}_0 \mid \mu \rangle, \tag{20}$$

where e_0 is the polarization of the observed light.

In the experiment, one measures the intensity of light with polarization $\hat{e}_0$; this is related to the quantities just mentioned as follows:

$$I(e_0) = K\sum_{nn'} A_{nn'}\rho_{nn'}. \tag{21}$$

We describe the nature of the observed signals for different forms of pumping. In the simplest case of interference phenomena and level crossing, the pumping is constant in time, all the remaining coefficients in (17) are also constant, and the differential equation for the density matrix (17) becomes an algebraic equation,

$$\frac{d}{dt}\rho_{nn'} = 0,$$
$$F_{nn'} - \Gamma_{nn'}\rho_{nn'} - i\omega_{nn'}\rho_{nn'} = 0, \tag{22}$$

whose solution has the form

$$\rho_{nn'} = F_{nn'}/(\Gamma_{nn'} + i\omega_{nn'}). \tag{23}$$

The observed intensity $I(e)$ is

$$I(e) = K\sum_{nn'} F_{nn'}A_{nn'}/(\Gamma_{nn'} + i\omega_{nn'}). \tag{24}$$

The expression (24) completely describes the signal. We split the sum in (24) into two parts, the first containing the terms with $n = n'$, the second containing those with $n \neq n'$. The first part characterizes the mean intensity of the observed signal:

$$\overline{I(e)} = K\sum_n A_{nn}F_{nn}/\Gamma_{nn}. \tag{25}$$

The second part characterizes the interference signal or, as one says, the interference intensity:

$$I_{\text{int}}(e) = K\sum_{n\neq n'} F_{nn'}A_{nn'}/(\Gamma_{nn'} + i\omega_{nn'}). \tag{26}$$

This term vanishes after averaging over all possible polarizations and directions of observation. The signal I_{int}, which arises because of the degeneracy of the Zeeman sublevels in a zero magnetic field, is called the Hanle signal (level crossing in zero magnetic field). The value of $\omega_{nn'}$ can be changed by means of an external magnetic field. Studying the dependence of I_{int} on the external magnetic field, $I_{\text{int}}(H)$, one can

determine the constants of the hyperfine interaction.

In the presence of a hyperfine interaction, because of the mixing of the states characterized in a zero magnetic field by the quantum numbers F_n and $F_{n'}$, the connection between $\omega_{nn'}$ and the value of the external magnetic field is determined in a fairly complicated manner by the parameters of the hyperfine structure, namely, by the constants of the magnetic dipole interaction of the electron shell with the nucleus; these are usually denoted by A, while the constant of the electric quadrupole interaction is denoted by B. The connection can be calculated by means of a computer, and as a result, given the magnetic field and the distance between the distinguished pairs of Zeeman sublevels, one can determine the constants A and B. The level-crossing phenomenon makes it possible to determine quite accurately from the signal I_{int} the value of the magnetic field at which the levels become degenerate ($\omega_{nn'} = 0$). These data serve to determine the constants A and B. Two measured points of intersection of different pairs of levels are, for a known Landé factor g_J, sufficient for the determination of both constants. This method is most promising in the case of determination of the constant B when all the remaining constants are known from independent measurements. On the basis of the values of the constants A and B and a calculation of the electron wave functions, one determines the magnetic dipole and electric quadrupole moments of the nucleus. One of the most important advantages of the method is that it does not require the exciting radiation to be highly monoenergetic but at the same time gives a high accuracy of the results. This means that one can work with radioactive elements in cells and poorly collimated beams of the nuclear reaction products.

It was assumed above that $F_{nn'}$ is constant. In the case of pulsed excitation and zero time delay between excitation and observation, the dependence $I_{\text{int}}(H)$ is the same as in the case of excitation by a cw laser. This means that in such experiments one can use not only cw dye lasers but also pulsed dye lasers. In addition, pulsed excitation may give a number of advantages.[55]

If the pumping depends periodically on the time with frequency Ω and on the modulation depth ε, i.e.,

$$F_{nn'}(t) = F^0_{nn'}[1 + (\varepsilon/2)\exp(i\Omega t) + (\varepsilon/2)\exp(-i\Omega t)],$$

then the solution of the differential equation (17) has the form

$$\rho_{nn'}(t) = F^0_{nn'}\left[\frac{1}{\Gamma_{nn'}+i\omega_{nn'}} + \frac{\varepsilon}{2}\left(\frac{\exp(i\Omega t)}{\Gamma_{nn'}+i(\omega_{nn'}+\Omega)}\right) + \left(\frac{\varepsilon}{2}\right)\frac{\exp(-i\Omega t)}{\Gamma_{nn'}+i(\omega_{nn'}-\Omega)}\right]. \quad (27)$$

The diagonal terms with $n = n'$, $\omega_{nn'} = 0$ give the signal

$$\bar{I} = K\sum_n F^0_{nn}A_{nn}\left[\frac{1}{\Gamma_{nn}} + \frac{\varepsilon}{2}\frac{\Gamma_{nn}[\exp(i\Omega t)+\exp(-i\Omega t)]}{\Gamma^2_{nn}+\Omega^2} + \frac{\varepsilon}{2}\frac{(i\Omega t)[\exp(i\Omega t)-\exp(-i\Omega t)]}{\Gamma^2_{nn}+\Omega^2}\right], \quad (28)$$

and taking the damping constants to be the same and equal to Γ, we obtain

$$\bar{I} = K\left[\frac{1}{\Gamma} + \frac{\varepsilon\cos(\Omega t-\varphi)}{\sqrt{\Gamma^2+\Omega^2}}\right]\sum_n F^0_{nn}A_{nn}, \quad (29)$$

where $\tan\varphi = \Omega/\Gamma$.

The fluorescence modulation depth $\varepsilon\Gamma/\sqrt{\Gamma^2+\Omega^2}$ is less than the pumping modulation depth and depends on the level lifetime $1/\Gamma$. The law of variation of the fluorescence intensity for modulated excitation has been used for measurement of the lifetime.[56]

For the interference term,

$$I_{\text{int}} = K\sum_{nn'} F^0_{nn'}A_{nn'}\left[\frac{2\Gamma_{nn'}}{\Gamma^2_{nn'}+\omega^2_{nn'}} + \frac{\varepsilon\cos(\Omega t-\varphi_1)}{\sqrt{\Gamma^2_{nn'}+(\omega_{nn'}+\Omega)^2}} + \frac{\varepsilon\cos(\Omega t+\varphi_2)}{\sqrt{\Gamma^2_{nn'}+(\omega_{nn'}-\Omega)^2}}\right], \quad (30)$$

where $\tan\varphi_1 = (\omega_{nn'}+\Omega)/\Gamma_{nn'}$ and $\tan\varphi_2 = (\omega_{nn'}-\Omega)/\Gamma_{nn'}$. The first term in the square brackets is the level-crossing effect; at high modulation frequencies, the second term is small, while the third term, the interference beats, reaches a maximum at $\omega_{nn'} = \Omega$.

If the difference $\omega_{nn'}$ between the resonance frequencies of the atom is equal to the modulation frequency of the external magnetic field, paramagnetic resonance occurs. The signal consists of an infinite series of resonances that decrease rapidly in amplitude. The width of the signal, as in all other interference phenomena, does not depend on the amplitude of the rf field.

In the case of pumping by a pulsed laser with short pulse duration and observation of the decay of the intensity in time, it is possible to detect characteristic beats in the radiation.[56] In this case, the pumping is represented by a δ function,

$$F(t) = F_0\,\delta(t-t_0), \quad (31)$$

where t_0 is the time at which the pulse is switched on.

The solution of Eq. (17) has the form

$$\rho_{nn'}(t) = F^0_{nn'}\exp[-(\Gamma_{nn'}+i\omega_{nn'})(t-t_0)]. \quad (32)$$

The fluorescence intensity is

$$I = \bar{I} + I_{\text{int}} = K\sum_n F^0_{nn}A_{nn}\exp[-\Gamma_{nn}(t-t_0)] + 2K\sum_{nn'}\exp[-\Gamma_{nn'}(t-t_0)]A_{nn'}F^0_{nn'}\cos\omega_{nn'}(t-t_0).$$

Signals corresponding to different variants of the experiments considered above are shown in Fig. 12.

Measurement of constants of the hyperfine interaction and determination of the quadrupole moments of nuclei from level-crossing signals

We consider in more detail experiments in which the level-crossing signals are observed, and we examine the method of determining the parameters A and B. In the experiments, one investigates the dependence of the fluorescence polarization, when atoms are irradiated by coherently polarized light, on the external magnetic field. The experiments can be done either with vapor of the substances in a cell or with atomic beams. We note, however, that only a small number of elements can be investigated in cells. Most elements interact chemically with the cell walls or require temperatures too high to produce a vapor density sufficient for observation of fluorescence. Therefore, most experiments have been done with atomic beams. The requirements on the atomic beam in level-crossing experiments are not the same as in spectroscopic investigations, since in the observation of level crossing the Doppler broadening associated

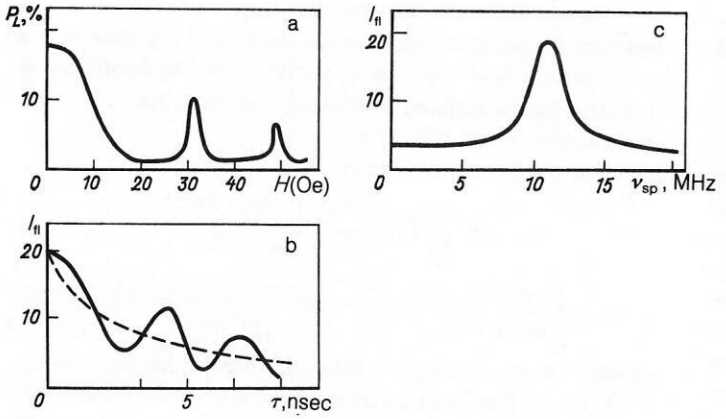

FIG. 12. Interference signals from coherent ensembles of atoms in the optical range: (a) level crossing; (b) beats in the radiation; (c) paramagnetic resonance (P_L is the degree of polarization of the fluorescence, and I_{fl} is the fluorescence intensity).

with the spread of the atom velocities is not important[53]; the divergence angle of the beam is limited by purely technical factors—it is required of the beam only that the windows through which the laser radiation passes and the observations are made remain transparent during the experiment, i.e., the investigated substance must not be precipitated on them. The remaining parameters of the experimental apparatus are the same as in the case when a cell is used.

In the case of irradiation with laser light, the strong field of the electromagnetic wave may influence the signal.[57] Familiarity with this form of signals, which themselves serve as a source of interesting information,[58] is necessary for the correct arrangement of the experiment and the interpretation of the experimental data. The distortions of this kind will be least if one chooses as the signal the difference between the intensities of the light with polarizations perpendicular to each other and to the magnetic field, the difference being divided by the sum of the intensities. Nevertheless, for accurate determination of the hyperfine interaction constants it is necessary to estimate the distortions introduced by the electromagnetic wave. For this, one makes measurements at several values of the power of the laser radiation.

As a rule, the most serious distortions are introduced in an experiment by capture of radiation; in addition, there may be errors associated with incorrect allowance for the spectral distribution of the intensities in both the irradiation and the observation channel. The remaining distortions are smaller than these.

The experimental data are analyzed on a computer by means of a special program. The calculations are made in two stages. In the first, the operator of the hyperfine interaction with the external magnetic field is diagonalized[59]:

$$V = A\,(\mathbf{IJ}) + B\left[\frac{3\,(\mathbf{IJ})^2 + (3/2)\,(\mathbf{IJ}) - I\,(I+1)\,J\,(J+1)}{2I\,(2I-1)\,J\,(2J-1)}\right] + g_J\mu_J\,(\mathbf{HJ}) - g_I\mu_I\,(\mathbf{HI}), \tag{33}$$

where A and B are the required constants of the hyperfine interaction, $\mathbf{I}$ is the spin of the nucleus, $\mathbf{J}$ is the spin of the electron shell, g_I and g_J are the gyromagnetic ratios for the nucleus and the electron shell, and H is the intensity of the external magnetic field.

The wave functions of both the ground and the excited state are calculated. The second step is the calculation of the signal $S(H)$:

$$S(H) = C \sum_{\mu,\, m,\, \mu',\, m'} \frac{\langle \mu \mid ed \mid m\rangle \langle m \mid e^* d \mid \mu'\rangle \langle \mu' \mid e' d \mid m'\rangle \langle m' \mid e'^* d \mid \mu\rangle}{(1/2)\,(\Gamma_\mu + \Gamma_{\mu'}) + i\,(E_\mu - E_{\mu'})/\hbar}, \tag{34}$$

where C is a constant determined by the geometry of the experiment, e is the polarization of the irradiating light, e' is the polarization of the detected radiation, and E and Γ are the energies and radiative widths of the intermediate (excited) states.

The time of the calculations for one set of constants is, in the case of overlapping signals, a few minutes. In the case of well-separated crossing signals the time of the calculations is greatly reduced, and in some cases the calculations can be made manually.

The level-crossing method is most often used to determine the quadrupole moments $\langle Q_0 \rangle$ of stable nuclei; in a number of cases, it has permitted the determination of the most accurate value of $\langle Q_0 \rangle$. For radioactive nuclei, such studies are as yet the only ones possible, and have been made for long-lived nuclides. Thus, the level-crossing method has been used to determine the quadrupole moments of the nuclei ^{131,132,136}Cs (Ref. 60), ^{107}Cd and ^{109}Cd (Ref. 61), ^{193m}Hg (Ref. 62), and ^{22}Na (Ref. 14).

We note that determination of the quadrupole moments from measurement of the hyperfine splitting makes it possible to obtain information directly about the value of $\langle Q_0 \rangle$, whereas various methods of nuclear physics make it possible to estimate $\langle Q_0^2 \rangle^{1/2}$. The values of $\langle Q_0 \rangle$ are insensitive to a number of dynamical changes in the shape of the nucleus, and in this sense the optical methods give information about the static nuclear quadrupole moment. The dynamical changes in the nuclear shape make a nonvanishing contribution to $\langle Q_0^2 \rangle^{1/2}$, i.e., $\langle Q_0^2 \rangle^{1/2}$ is a certain sum of the static and dynamical quantities. Differences between the values of $\langle Q_0 \rangle$ and $\langle Q_0^2 \rangle^{1/2}$ were noted in Refs. 5 and 63 for a number of ground and isomer states of nuclei. Attempts were made

to explain these differences by the existence of dynamical changes in the shape of the nuclei, including zero-point fluctuations of the shape.

COHERENT EFFECTS IN NUCLEAR SYSTEMS

When an atom is irradiated with light of coherent polarization, the presence of the hyperfine interaction has the consequence that there is coherence in not only the electron shell but also, in a number of cases, nuclear coherence as well, i.e., the nuclear substates differing in the magnetic quantum number are connected in phase. In this section, we consider questions associated with the mechanism of occurrence of nuclear coherence, with the types of optical transitions whose excitation leads to the appearance of nuclear coherence, with the details of the radioactive decay of nuclei in such systems, and with the experimental investigation of the manifestation of nuclear coherence in the optical and nuclear radiations. We shall give the description in the density-matrix formalism.

Manifestation of the nuclear coherence in the optical radiation

We begin by separating from the density matrix of the atom the elements of the density matrix of the nucleus and the electron shell. For this, we make a tensor decomposition of the density matrix of the atom into the density matrix of the nucleus and the electron shell. The method of such decompositions is explained in Ref. 52. Let $|Fm_F\rangle$ be the states of the atom and ρ_F the density matrix; $|Im_I\rangle$ be the states of the nucleus and ρ_I the density matrix; and $|Jm_J\rangle$ be the states of the electron shell and ρ_J the density matrix. Since the total angular momentum of the atom is the vector sum of the angular momenta of the nucleus and the electron shell, the density operator ρ_F is a tensor product of first rank of the operators ρ_I and ρ_J:

$$\rho_F = \rho_I \otimes \rho_J, \tag{35}$$

or

$$\langle Fm_F \mid \rho_F \mid F'm'_F\rangle = \sum G\,(IFF'JJ'I')\,\langle Im_I \mid \rho_I \mid I'm'_I\rangle \langle Jm_J \mid \rho_J \mid J'm'_J\rangle\,\langle I'm'_IJ'm'_J \mid F'm'_F\rangle\,\langle Jm_JIm_I \mid Fm_F\rangle. \tag{36}$$

This and a number of other relations can be obtained by using the graphical technique.[52] For the components of the density matrix, after decomposition with respect to irreducible tensor operators, we obtain ($I = I'$, $J = J'$)

$$\rho_{kq}(F, F') = \sum_{k_1q_1k_2q_2} \rho_{k_1q_1}(I)\,\rho_{k_2q_2}(J)\,\langle k_1q_1k_2q_2 \mid kq\rangle \times \hat{F}\hat{F}'\hat{k}_1\hat{k}_2 \begin{Bmatrix} I & J & F \\ I & J & F' \\ k_1 & k_2 & k \end{Bmatrix}, \tag{37}$$

where $\hat{x} = \sqrt{2x+1}$.

In the case of excitation of only one of the hyperfine components of the atomic state, we can set $F = F'$, $I = I'$, $J = J'$; then

$$\langle Fm_F \mid \rho_F \mid Fm'_F\rangle = \sum G\,(F,\ I,\ J)\,\langle Im'_IJm'_J \mid Fm'_F\rangle \times \langle Jm_JIm_I \mid Fm_F\rangle\,\langle Im_I \mid \rho_I \mid Im'_J\rangle\,\langle Jm_J \mid \rho_J \mid Jm'_J\rangle. \tag{38}$$

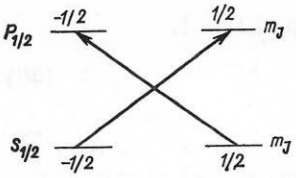

FIG. 13. Scheme of excitation of a transition in the ^{23}Na atom by a broad line.

It follows from the expression (38) that if ρ_I and ρ_J are diagonal, then so is ρ_F. Indeed, diagonality of ρ_I and ρ_J means that the only nonvanishing elements of the density matrix are the ones for which $m_I = m'_I$, $m_J = m'_J$, and this, in its turn, means that $m_F = m'_F$, i.e., ρ_F is diagonal. Generally speaking, in the presence of coherence in the atom (nondiagonality of ρ_F) the presence of coherent nuclear states is not necessary, since the matrix ρ_F can have nonzero nondiagonal elements if the matrix ρ_J is nondiagonal. However, in a number of cases a necessary condition for nondiagonality of ρ_F is nondiagonality of ρ_I. In such cases, investigation of the interference effects in the optical radiation of the atom associated with the nondiagonality of ρ_F makes it possible to draw conclusions about the nondiagonal elements of the matrix ρ_I. This is particularly important when one is planning experiments to study interference phenomena in nuclear radiation that are associated with nondiagonality of ρ_I. One of these cases is the effect of the occurrence of $\Delta m = 2$ coherence when Na vapor is excited by linearly polarized light in the D_1 line. In Ref. 14, experiments with a ^{23}Na atomic beam were made. In the case of excitation by a broad line, i.e., without separation of the hyperfine transition components, coherence does not arise. For although the selection rules do permit transitions with $\Delta m = \pm 1$ from a given magnetic sublevel of the ground state, there is no coherence because the excited state does not have sublevels differing in the magnetic quantum number by $\Delta m = 2$ (Fig. 13). A quite different situation is obtained when we use a narrow line for the excitation. In this case, since there are states differing in the magnetic quantum number by $\Delta m = 2$, coherence arises in the system (Fig. 14).

As an example, we consider excitation of the transition $F = 1$, $S_{1/2} \to F = 1$, $P_{1/2}$. The density matrix $\rho_{m_Fm'_F}$ of the excited state of the atom has the form

$$\begin{pmatrix} \rho_{-1-1} & 0 & \rho_{-11} \\ 0 & \rho_{00} & 0 \\ \rho_{1-1} & 0 & \rho_{11} \end{pmatrix}, \tag{39}$$

where

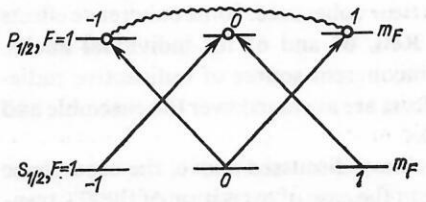

FIG. 14. Scheme of excitation of a transition in the ^{23}Na atom by a narrow line. The wavy line connects coherent states.

$$\rho_{1-1} = \langle F = 1,\ m_F = 1 \mid \rho_F \mid F = 1,$$
$$m_F = -1\rangle,\ \rho_{1-1} = \rho^*_{-11}, \tag{40}$$

or, using (38),

$$\rho_{1-1} = {}^{1}\!/_{6}\ \langle 3/2\ -3/2\ 1/2\ \ 1/2 \mid 1\ -1\rangle \times \langle 1/2\ -1/2\ 3/2\ 3/2 \mid 11\rangle \times \langle 3/2\ {}^{3}\!/_{2} \mid \rho_I \mid 3/2\ -3/2\rangle\ \langle 1/2\ 1/2 \mid \rho_J \mid 1/2\ -1/2\rangle. \tag{41}$$

It follows from the expression (41) that if the matrix ρ_I is diagonal, then so too will be ρ_F, while if ρ_F is nondiagonal, so too will be ρ_I.

If the transition $F = 1, S_{1/2} \to F = 2, P_{1/2}$ is excited (see Fig. 1), then the density matrix of the atom has the more complicated form

$$\begin{Bmatrix} \rho_{-2-2} & 0 & \rho_{20} & 0 & 0 \\ 0 & \rho_{-1-1} & 0 & \rho_{-11} & 0 \\ \rho_{0-2} & 0 & \rho_{00} & 0 & \rho_{02} \\ 0 & \rho_{1-1} & 0 & \rho_{11} & 0 \\ 0 & 0 & \rho_{20} & 0 & \rho_{22} \end{Bmatrix}. \tag{42}$$

Using the relations (36) and (38), we can in this case too readily conclude that if ρ_F is nondiagonal, then so is ρ_I, and vice versa.

In the optical radiation, as noted earlier, an interference signal occurs if ρ_F is nondiagonal:

$$I = K \frac{\sum \langle f \mid \hat{D} \mid i\rangle \rho_{ij} \langle j \mid \hat{D} \mid f\rangle}{\sum \langle f \mid \hat{D} \mid i\rangle \rho_{ii} \langle i \mid \hat{D} \mid f\rangle}. \tag{43}$$

In Ref. 14, the interference signal was investigated for excitation of ^{23}Na atoms by a narrow line. In the observation channels, the components of the hyperfine structure were also separated and the interference signal was studied. In Ref. 14, an estimate was made of the degree of nuclear coherence

$$K = |\rho^I_{m_1 m_2}|^2 / |\rho^I_{mm}|^2.$$

It was found to be $K \sim 0.11$. Here, $|\rho^I_{m_1 m_2}|^2$ is the square of the amplitude of the nondiagonal element of the density matrix of the nucleus responsible for the observed effect, and $|\rho^I_{mm}|^2$ is the mean value of the square of the amplitude of the diagonal element of the nuclear density matrix.

The coherence of nuclear sublevels may lead to a number of interference phenomena in the nuclear radiation: beats, crossing of the levels of the atom in weak magnetic fields as well as of the nucleus in strong fields, and polarization of the radiation. It should be noted that in the literature questions related to the use of a coherent nuclear source of radioactive radiations have not been analyzed, so that at the present time it is only possible to outline the main features of the application of nuclear coherence. Some coherence effects were considered in Refs. 64 and 65 for individual nuclei. However, when an incoherent source of radioactive radiations is used these effects are averaged over the ensemble and become unobservable.

Besides what we have discussed above, the occurrence of nuclear coherence in the case of excitation of the D_2 transition in sodium was analyzed in Ref. 14. It was shown that in this case nuclear coherence arises if the system is illuminated by a broad line, and this, in its turn, makes it possible to observe a number of interference phenomena without separating the components of the hyperfine structure.

Observation of the level-crossing phenomenon in nuclear radiation in strong magnetic fields

In the case of radioactive decay of nuclear systems that possess coherence, an interference signal must be observed in the nuclear radiation:

$$I_{\text{int}}(H) = \frac{\sum \langle f \mid \hat{D} \mid i\rangle \rho^I_{ij} \langle j \mid \hat{D} \mid f\rangle}{\sum \langle f \mid \hat{D} \mid i\rangle \rho^I_{ii} \langle i \mid \hat{D} \mid f\rangle}, \tag{44}$$

where ρ^I is the density matrix for the ensemble of nuclei, and $\hat{D}$ is the operator of the nuclear transition.

Superposition states in which different nuclear substates are connected in phase can be excited either by means of laser radiation or directly in accelerator beams by irradiating the target with protons, α particles, etc. In the first case, the nuclear radiation will manifest the crossing of the hyperfine-structure levels in the atoms, and observation of the crossing signals requires weak magnetic fields (about 100 Oe). In the second case, crossings of the levels of the nuclei will be directly observed, and magnetic fields of several tens of kilo-oersteds are then needed.

In Ref. 66, the crossing of nuclear levels in ^{69}Ge was studied through the γ radiation. A study was made of the isomer state in ^{69}Ge with $I^\pi = 9/2^+$, $E = 398$ keV, $\tau = 4$ μsec. The isomer was populated by irradiating a zinc crystal with 20-MeV α particles. The intensity of the γ radiation of the $M2$ transition $9/2^+ \to 5/2^+$ was investigated at definite angles as a function of the external magnetic field. The energy of the nuclear Zeeman sublevel in the crystal is

$$E_m = \hbar\,(3\ m^2\omega_Q\ + m\omega_L\ + \text{const}), \tag{45}$$

where

$$\omega_L = -\,g\mu H/\hbar,$$
$$\omega_Q = \omega_0/3 \text{ (integral } I\text{) or } 6\omega_Q \text{ (half-integral } I\text{)},$$
$$\omega_Q = eQV_{zz}\ \ [4I\ (2I\ -\ 1)/\hbar].$$

Here, H is the magnetic field strength, V_{zz} is the gradient of the electric field, Q is the quadrupole moment of the nucleus, and g is the gyromagnetic ratio.

Resonance in the γ radiation occurs for

$$\omega_L/\omega_0 = \tfrac{1}{2}\ n\ \ (n = 1,\ 3,\ 5\ \ldots),$$

when two levels with $\Delta m = 2$, etc., cross. An experimental study was made of the Lorentz and dispersion shapes of the signals:

$$R_L(H) = \frac{W(0°) - W(90°)}{W(0°) + W(90°)}, \tag{46}$$

$$R_D(H) = \frac{W(45°) - W(135°)}{W(45°) + W(135°)}, \tag{47}$$

where $W(\theta)$ is the counting rate of the γ rays at angle θ to the direction of the α-particle beam. The dependences of the Lorentz, R_L, and dispersion, R_D, signal shapes are shown in Fig. 15. The first crossing was observed in a field $B \sim 0.94$ T. From the results of the experiments, the authors of Ref. 66 obtained estimates of the gradient of the electric field in the zinc crystal and a number of constants of relaxation processes.

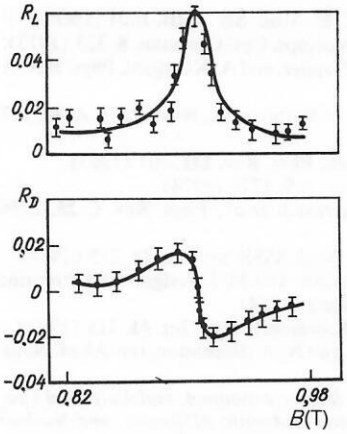

FIG. 15. Lorentz and dispersion shapes of the crossing signal observed in the γ radiation.

The method described above can be used in experiments on level anticrossing. Such experiments augment the information obtained in level-crossing experiments.[53]

CONCLUSIONS

With the appearance of lasers with tunable frequency it became possible to develop several very original and effective methods for investigating atoms, molecules, and nuclei by means of laser radiation. It is important that in many cases the use of laser radiation makes it possible to solve a problem not hitherto solved or to radically simplify or improve an existing method of investigation.[68] The high sensitivity and resolution of the methods of laser spectroscopy make it possible to investigate characteristics of nuclei available in small numbers, and the possibility of smooth tuning of the frequency of the laser radiation opens up wide possibilities for working with atoms and molecules containing radioactive nuclei and for investigating the properties of these nuclei. However, in the investigation of the properties of nuclei so far made by means of laser radiation only two of its properties have generally been used—its highly monoenergetic nature and the high intensity. The coherence property of laser radiation has not yet been widely used in nuclear-physics investigations.

In the present review, we have considered the possibilities of laser technology for the solution of problems associated with obtaining polarized nuclei and nuclei with polarization coherence. However, these are only a small fraction of the methods developed in coherent optics that could be used in nuclear-physics investigations. The development and application of the methods of coherent optics to nuclear physics is a very interesting direction of research.

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