Rapid nuclear polarization by pulsed lasers

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Nuclear orientation by means of a short (10^{-8} sec duration) pulse of circularly polarized light is investigated. The source of radiation was a tunable dye laser with pulse power up to 15 kW. The anisotropy in the angular distribution of the γ rays emitted by the radioactive ^{22}Na and ^{24}Na nuclei following irradiation of sodium vapors by 589.6 nm laser light was studied. A degree of nuclear polarization of about 10% was achieved in a time less than the duration of the laser pulse.

INTRODUCTION

In experiments involving oriented nuclei an important role is played by optical polarization methods.[1] In recent years these methods have been further developed by employing tunable lasers. In the optical pumping method,[2] when the atoms interacting with the circularly polarized light are all put into the state with maximum projection of angular momentum along the direction of the laser beam, the degree of nuclear polarization can reach a 100%. However such a polarization is achieved in a time corresponding to several spontaneous transitions from the excited to the ground states of the atom (10^{-7}–10^{-6} sec). This is a limitation on the study of nuclei in isomer states with shorter lifetimes.

For the polarization of such short-lived nuclei the method of Rabi oscillations[3,4] or π-pulse[5] may be used. In these methods the nonequilibrium population of states with different values for the projection of total angular momentum is achieved only as a result of induced transitions, caused by the circularly polarized radiation. Therefore the limiting lifetimes of the nuclei under study are determined by the magnitude of the hyperfine interaction between the electron shell and the nucleus and may amount to $\sim 10^{-10}$ sec, although in that case the degree of polarization does not reach 100% (it turns out to be 1–50%, depending on the spins of the nucleus and the electron shell). The nuclear polarization processes for optical pumping and for Rabi oscillations are shown schematically in Fig. 1.

The frequency of Rabi oscillations is determined by the power density of the laser radiation which should be substantial (in excess of 10^3 W/cm^2) if rapid nuclear polarization is to be achieved. Such power is obtained most readily from pulsed lasers. The aim of this paper is an investigation of the polarization mechanism of the radioactive ^{22}Na and ^{24}Na and the stable ^{23}Na nuclei by means of tunable pulsed lasers. The polarization of these nuclei was determined by measuring the angular anisotropy of the γ rays emitted in their decay or the polarization of the resonant fluorescent light radiation.

EXPERIMENTAL SETUP

A schematic of the experimental setup, used in the study of effects of orientation of the radioactive nuclei, is shown in Fig. 2. The source of the light pulse was a tunable dye laser—rhodamine B. The change in the wavelength between the limits of 575–600 nm was accomplished by smooth rotation of the diffraction grating. The width of the generating line amounted to 0.25 Å (20 GHz). The circular polarization of the radiation was produced by a linear polarizer and a quarter-wavelength plate, rotated relative to the plane of linear polarization of the laser beam by an angle of $\pi/4$.

The pulse excitation of the dye was accomplished by a Q-switched solid-state laser based on a single crystal of aluminum-yttrium garnet alloyed with neodymium (LTIPCh-7). This laser generates pulses of light of 1064 nm wavelength and 10 nsec duration, with up to 1 MW power per pulse and pulse repetition frequency from 12.5 to 100 Hz. To excite the dye, a transformation of the radiation wavelength to the second harmonic (with $\lambda = 532$ nm) was carried out with the help of a lithium niobate nonlinear crystal. The power of the laser radiation at the dye amounted to 15 kW (for the same values of pulse frequency and duration).

Resonant excitation of the atoms under study was performed in a cell of ~ 1 cm^3 volume made out of "kaprolon"—a material with small desorientation cross section for Na atoms, transparent to optical radiation, and ensuring sufficiently high vacuum. The cell was located inside a vacuum chamber, pumped out to a pressure of 10^{-6} Torr, which could be heated to a temperature of 400° C by means of an electric current passing through a coil. For the atomization of the various compounds, powerful pulsed laser radiation was used (from that same LTIPCh-7 laser).

To eliminate atom desorientation due to random weak fields a sufficiently strong external magnetic field (up to 200 G), produced by a pair of Helmholtz coils, was applied in the direction of the laser beam.

Detection of the γ radiation, emitted in the radioactive decay of oriented nuclei, was performed by a scintillation

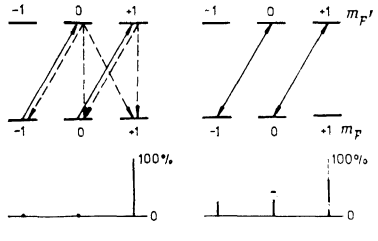

FIG. 1. The process of nuclear polarization in optical pumping (left) and in Rabi oscillations (right). Solid arrows—induced transitions, dashed—spontaneous transitions, m_F—projection of the total angular momentum of the atom in the direction of the laser beam. Below—relative populations of states with different m_F.

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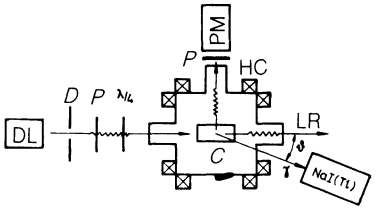

FIG. 2. Block diagram of the setup. DL—dye laser, *D*—diaphragm, *P*—linear polarizer, $\lambda/4$—quarter wave plate, *C*—cell, HC—Helmholtz coils, LR—laser radiation.

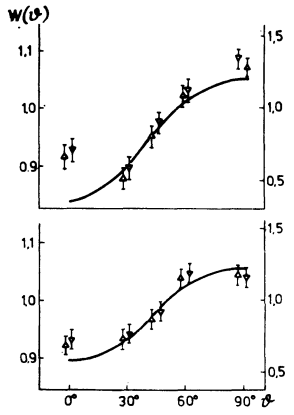

FIG. 3. Angular distribution of the γ radiation from the ^{22}Na (top) and ^{24}Na (bottom) nuclei. The dots and the left ordinate scale refer to the experiment, the solid curve and the right scale refer to calculations from Eq. (1) with corrections for the angular resolution of the detector.

spectrometer with a NaI(Tl) crystal of 60×60 mm dimension. The detector could be placed at various angles (from 0° to 90°) relative to the laser radiation and it subtended a solid angle of 0.28 sr. The spectra of the γ radiation at various angles and directions of the magnetic field were registered by a multichannel analyzer.

The chamber had glass windows to allow the transmission of the laser radiation, as well as of resonant fluorescent radiation when working with the stable isotopes. The latter was detected by a photomultiplier preceded by a rotating polaroid. This photomultiplier and polaroid made possible measurements of the polarization of the resonant scattered radiation and thus judge the orientation of the atoms.

POLARIZATION OF ^{22}Na AND ^{24}Na NUCLEI

Experiments were performed with the described setup to measure the angular anisotropy of the γ radiation from ^{22}Na and ^{24}Na, arising from their polarization under the action of the pulsed laser radiation. The activity of the sources amounted to 3–5 μCi, corresponding to $\sim 10^{13}$ nuclei for ^{22}Na and $\sim 10^{10}$ nuclei for ^{24}Na. The characteristics of the γ radiation (its energy and spin sequences in the γ-decay and the preceding β-decay[6]) are given in Table I.

The sources, in the form of a NaCl salt, were deposited on a tantalum substrate and placed in a cell which, following evacuation, was irradiated by the pulsed laser for 8–10 seconds. In the process the salt would evaporate and decompose so that no less than 70% of the evaporated sodium was in atomic state.[7] The tray was heated to a temperature of 200° C, at which effects of desorientation of the atoms on the walls are minimal. In addition, at this temperature the pressure of the saturated sodium vapor is such that for the above-indicated number of atoms with radioactive nuclei there is no condensation of them on the walls.

The layer of Na vapor in the cell, obtained in this manner, was irradiated by circularly polarized laser radiation of 589.6 nm wavelength, corresponding to the D1 line of Na (transition from the ground $S_{1/2}$ to the first excited $P_{1/2}$ state). The diameter of the laser beam corresponded to the size of the cell so that a major fraction of sodium atoms was in the excitation zone.

Measurements on the γ radiation spectra of ^{22}Na and ^{24}Na were performed at 0°, 30°, 45°, 60° and 90° angles relative to the direction of the laser beam. At each angle four γ-spectra were measured, corresponding to two opposite directions of the magnetic field (along the laser ray and opposite) with the laser beam turned on and off. Such a method permits the elimination of corrections due to changes in the solid angle and detection efficiency of the γ radiation, as well as corrections related to the effect of the magnetic field on the photomultiplier. By comparing the intensity of the γ radiation belonging to ^{22}Na or ^{24}Na, in the presence and absence of the laser ray, one may obtain the angular distribution of the γ radiation of nuclei that had been oriented by the polarized laser light. This angular distribution is shown in Fig. 3, where for comparison we also show the angular distribution of γ radiation of oriented nuclei, calculated from the familiar expression[8]

$$w(\vartheta)=1+A_2P_2(\cos\vartheta)+A_4P_4(\cos\vartheta). \qquad (1)$$

Here $P_2(\cos\vartheta)$ and $P_4(\cos\vartheta)$ are Legendre polynomials, and A_2 and A_4 are angular correlation coefficients that depend on the spins of the levels and the multipolarity of the transitions (they are given in Table I for the γ transitions in ^{22}Na and ^{24}Na).

It is seen from Fig. 3 that the angular distribution of the γ radiation is anisotropic, indicating nuclear orientation under the action of the circularly polarized laser radiation. However the magnitude of the anisotropy (the relative excess of γ radiation intensity at 90°), amounting to 15% and

TABLE I. Nuclear characteristics of sodium isotopes.

Nucleus	*E*, keV	$I_i \to I_f$		A_2	A_4
		β-decay	γ-decay		
^{22}Na	1280	$3^+ \to 2^+$	$2^+ \to 0^+$	−0.714	−0.286
^{24}Na	2753	$4^+ \to 4^+$	$4^+ \to 2^+$	−0.608	−0.143

12% respectively for ^{22}Na and ^{24}Na, is noticeably less than what is calculated with the help of Eq. (1). This means that not all nuclei are polarized at the instant of emission of the photon. In that case the expression for the angular distribution has the form:

$$W(\vartheta)=1+P_I A_2 f_2 P_2(\cos\vartheta)+P_I A_4 f_4 P_4(\cos\vartheta), \qquad (2)$$

where P_I is the degree of polarization of the nuclei, and f_k is the polarization attenuation in the β-decay preceding the emission of the photon. The degree of polarization of a nucleus with spin I is the determined by the average of the spin projection on the direction of the laser radiation I_z or the relative population of the corresponding magnetic sublevel n_m:

$$P_I=|\langle I_z\rangle/I|=\left|\sum_m m_I n_m \Big/ I\sum_m n_m\right|. \qquad (3)$$

The attenuation coefficient f_k is determined by the ratio of degrees of polarization of the intermediate (from which the photon emission proceeds) and initial states of the nucleus. If all nuclei are in the state with maximum spin projection ($n_m(I)=\delta_{mm'}$) then the expression for f_k has the form[9]

$$f_k=\binom{2k}{k}^{-1} I^{-k}\frac{(2I)!}{(2I-k)!}. \qquad (4)$$

In the case of ^{22}Na there is the polarization attenuation by the β-decay, while for ^{24}Na we have the values $f_2=0.85$ and $f_4=0.50$. The measured values of the anisotropy of the γ radiation and the calculated values of f_k permit a determination of the degree of polarization of the nuclei under study. It amounts to $(12\pm1)\%$ for ^{22}Na and $(8\pm1)\%$ for ^{24}Na. It is necessary to note that this degree of polarization is averaged over the entire time interval between laser pulses.

DISCUSSION OF RESULTS

We consider in more detail the process of polarization of nuclei by a short laser pulse, when its duration (10^{-8} sec) is less than the time for spontaneous relaxation of the electron shell (the lifetime of the excited level of the sodium atom is $1.6\cdot10^{-8}$ sec). Values of isotopic shifts for ^{22}Na and ^{24}Na relative to the stable ^{23}Na, as well as the hyperfine splitting for the ground and excited states, are given in Table II.[10]

It is seen from Table II that for a width of the laser excitation line of 20 GHz complete coverage of all isotopic shifts and components of hyperfine structure is ensured, as well as of the Doppler contour ($\sim$2 GHz) of the Na atoms in the cell. Therefore for an energy of the laser pulse of 0.15 mJ ($\sim10^{15}$ photons) saturation of the atomic transition takes place, i.e., in the time of the laser pulser (10^{-8} sec) practically all atoms that fell into the irradiation zone experience resonant excitation and the Rabi oscillation regime may be realized.

In the Rabi oscillation regime the degree of nuclear polarization (in percent) is determined by the expression

$$P_I=\frac{100}{(2I+1)(2J+1)}\sum_{m_F}\mathscr{P}_{m_F}, \qquad (5)$$

where I and J are the spins of the nucleus and electron shell respectively, F is the total angular momentum of the atom, m_F is its projection in the direction of the laser beam, and

TABLE II. Optical characteristics of sodium isotopes.

Isotope	$\Delta\nu_{is}^{23,A}$, MHz	$\Delta\nu_{hfs}(S_{1/2})$, MHz	$\Delta\nu_{hfs}(P_{1/2})$, MHz
^{22}Na	−754	1220	130
^{23}Na	0	1772	190
^{24}Na	+705	1140	126

$\mathscr{P}_{m_F}$ is the polarization coefficient. An expression for $\mathscr{P}_{m_F}$ and its actual magnitude for various values of I, J, and F were obtained in Ref. 11. In Fig. 4 we show the dependence of the degree of polarization of the ^{22}Na nucleus on the parameter Z, which is determined by the frequency of Rabi oscillations $\Omega(t)$:

$$Z=\int_0^\tau \Omega(t)\,dt, \qquad (6)$$

where τ is the duration of the laser pulse. The frequency of the Rabi oscillations depends on the magnitude of the matrix element of the atomic transition $V(J)$ and on the power W of the laser pulse:

$$\Omega\sim V(J)W^{1/2}. \qquad (7)$$

It can be seen from Fig. 4 that the dependence of the degree of polarization on the power of the laser radiation is an oscillatory function. A similar dependence holds for the ^{24}Na nucleus. The maximum in this dependence, when the degree of polarization amounts to $\sim12\%$, corresponds to a power of laser radiation for which in the time of a radiation pulse only one induced transition to the excited state of the atom takes place (π-pulse regime). It was at a power close to that required to achieve the π-pulse regime that we performed the measurements of the angular distribution of the γ radiation.

Spontaneous transition from the excited state of the atom to the ground state proceeds already after the laser pulse is over. In this process the degree of nuclear polarization is decreased, but it is restored to a higher value during the following laser pulse. In several laser pulses one may achieve, in principle, a degree of polarization close to a 100%, as in optical pumping. However, under realistic conditions this does not happen, owing to depolarization on the walls of the cell discussed above and to collisions with impurity atoms. In our experiments the time between pulses of

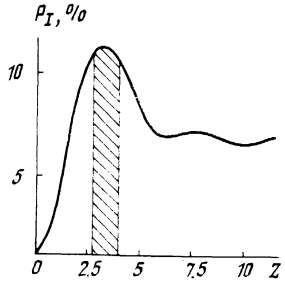

FIG. 4. Dependence of the degree of nuclear polarization of ^{22}Na on the parameter Z defined by Eq. (6). The dashed region corresponds to the intensity of the laser radiation used in the experiments.

laser radiation amounted to 80 msec, so that effects of depolarization could be significant.

It is the depolarization of the Na atoms, as well as the fact that not all of them experience resonant excitation (some of the atoms are adsorbed by the walls of the cell or are in the form of a compound) which explains why the observed degree of polarization of the ^{22}Na and ^{24}Na nuclei is substantially less than a 100% and close to what would be calculated for a single laser pulse. Apparently, the increase in the degree of nuclear polarization with each laser pulse is compensated by the depolarization between pulses (the depolarization time should amount to tenths of a second).

This mechanism for nuclear orientation is confirmed by measurements of angular anisotropy and polarization of resonant fluorescent radiation, as well as by the dependence of these quantities on the power of the laser radiation, in the case of the stable isotope ^{23}Na.[12] These measurements made possible the determination of the degree of orientation of the atom after the π-pulse and after spontaneous relaxation of the electron shell, hence also the uniquely determined by them degree of polarization of the nucleus, in good agreement with calculations from the formulas, Eqs. (5) and (6).

In this fashion these measurements and calculations show that pulsed circularly polarized laser radiation permits the achievement of a degree of nuclear polarization sufficient for experiments with radioactive nuclides. The polarization was accomplished in one laser pulse of short duration and high power in a time up to 10^{-10} sec, which opens up prospects for studies of short-lived nuclei.

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