Polarization of ^{23}Na atoms and nuclei by pulsed laser radiation

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The mechanism of orientation of ^{23}Na atoms and nuclei by short (10^{-8} s) pulses of polarized light is investigated. A tunable dye laser of pulse power up to 15 kW is used. The dependence of the degree of polarization of the resonance fluorescence due to the D 1- and D 2-transitions in ^{23}Na on the scattering angle and laser irradiation power is measured. Up to 35% of the atoms and 20% of the nuclei are polarized in a time shorter than the duration of the laser pulse.

INTRODUCTION

The interaction of polarized optical radiation with atoms leads to the efficient orientation of nuclei thanks to the hyperfine interaction between the electron shell and the nucleus.[1] In the optical pumping method[2] the degree of polarization can reach 100%, but this is realized after a time corresponding to some spontaneous transitions from an excited state of the atom to the ground state (10^{-7}–10^{-6}s). In Ref. 3 it was shown that in the presence of circularly polarized radiation of sufficiently strong power Rabi oscillations arise (induced transitions between the ground level and the excited level of the atom) which lead to a nonequilibrium population of the atomic states with different values of the projection of the total angular momentum. Even though the degree of polarization of the nucleus in this case is less (1–50%, depending on the spin of the nucleus and of the electron shell), the time necessary for polarization is no longer connected with the relaxation of the excitation, but is determined by the magnitude of the hyperfine splitting and can reach 10^{-10} s. Such an approach makes it possible to realize efficient polarization of short-lived nuclei with lifetimes down to fractions of a nanosecond.

The possibility of polarization of nuclei by a short (duration of 10^{-8} s) pulse of laser radiation was demonstrated in our previous paper[4] in the instance of the radioactive nuclei ^{22}Na and ^{24}Na. Upon irradiation of sodium atoms in the gas phase a degree of nuclear polarization of $\sim 10\%$ was achieved, measured by determining the anisotropy of the γ radiation of the indicated nuclides. However, this polarization, averaged over the time interval between the laser pulses (80 ms), significantly depends on depolarization effects and therefore does not allow one to make detailed judgments on the process of orientation of the nucleus during the interaction of the atom with the optical radiation.

To better understand the process of orientation of the nuclei, it is of interest to measure their degree of polarization immediately after the laser pulse. These measurements can be carried out in two ways.

1. The direct approach, consisting of measuring the angular anisotropy of the radioactive radiation of the short-lived isomer with lifetime comparable with the duration of the laser pulse.

2. The indirect approach, connected with the measurement of the angular distribution and polarization of the optical radiation of the excited atoms. The hyperfine interaction between the electron shell and the nucleus leads to the orientation of the nucleus, and this causes depolarization and smoothing of the angular distribution of the fluorescence radiation of the excited atoms.[5] Therefore measurements of these quantities also allow one to judge the degree of polarization of the nuclei during the optical excitation of the atoms.

The measurement of the polarization and angular distribution of the resonance fluorescence radiation of ^{23}Na atoms excited by short pulses of polarized laser radiation to the states $p_{1/2}$ and $p_{3/2}$ (the optical transitions D 1 and D 2) is the aim of the present work.

ANGULAR DISTRIBUTION AND POLARIZATION OF THE FLUORESCENCE RADIATION

The magnitudes of the hyperfine splitting in the $p_{1/2}$ (190 MHz) and $p_{3/2}$ states (16–60 MHz) correspond to times of the order of 10^{-9} s. These times are shorter than the lifetimes of the excited Na atoms spontaneously decaying to the ground state ($1.6\cdot 10^{-8}$ s). Therefore the polarization of the nucleus can take place also during its interaction with the excited electron shell of the atom.

Let us consider the process of orientation of the atom and the nucleus during the absorption of linearly or circularly polarized photons and analyze the angular distribution and polarization of the fluorescence during spontaneous emission to the ground state. In this case the quantization axis and the external magnetic field are directed along the laser beam in the case of circularly polarized radiation and along the polarization vector of the light in the case of linear polarization. The atom is characterized by its total angular momentum F, which is the vector sum of the angular momenta of the nucleus I and the electron shell J, and also by its projection M on the quantization axis. The selection rules for the magnetic quantum numbers allows a transition from any sublevel of the ground state $|FM\rangle$ of the atom to not more than one sublevel of the excited state $|F^*M^*\rangle$, and the density matrix of the excited state is diagonal with respect to the magnetic quantum numbers:

$$\rho_{MM^*}=\delta_{MM^*}\rho_M. \tag{1}$$

This matrix is expressed in terms of the irreducible tensor operators,[6] whose rank for the level with total angular momentum T_{kq} and for dipole absoprtion of a photon with spin $s=1$ is bounded:

$$k\leqslant F, \qquad k\leqslant 2s\,. \tag{2}$$

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Here $k = 1$ corresponds to polarization and $k = 2$ to alignment of the spins of the atoms (or nuclei). Explicit expressions for the tensor operators are given in Ref. 7. The system of atoms with aligned spins is characterized by the parameters A_0 and A_2, which are proportional to the values of the operators $\langle T_{00} \rangle$ and $\langle T_{20} \rangle$ averaged over the components of the hyperfine structure (the value of $\langle \overline{T}_{00} \rangle$ equals 1 in all cases):

$$A_2^F \equiv \langle \overline{T}_{20} \rangle = \sum_{F,M} \frac{\langle FM | 3F_z^2 - F^2 | FM \rangle}{F(F+1)}. \tag{3}$$

An analogous parameter P_F is introduced for the system of polarized atoms:

$$P_F \equiv \langle \overline{T}_{10} \rangle = \sum_{F,M} \frac{\langle FM | F_z | FM \rangle}{F}. \tag{4}$$

The fluorescence intensity at the angle ϑ with respect to the direction of the laser beam with given polarization is expressed in terms of these parameters:[8]

$$I_f(\vartheta, \beta, \psi) = c[1 + {}^1/_4 A_2^F (1 - 3\cos^2\vartheta + 3\sin^2\vartheta \cos 2\beta \cos 2\psi) + {}^3/_2 P_F \cos\vartheta \sin 2\beta \cos 2\psi], \tag{5}$$

where c is a normalizing factor, the angle β determines the polarization of the laser radiation ($\beta = 0$ for linear and $\beta = \pm \pi/4$ for circular polarization), and the angle ψ determines the polarization of the fluorescence. For circular polarization $\psi = 0$, and for linear polarization $\psi = 0$ if the polarization vector lies in the plane of the incident and resonant scattered radiation ($I_f = I_\parallel$) and $\psi = \pi/2$ if the polarization vector is perpendicular to this plane ($I_f = I_\perp$). These directions of the vectors with respect to the plane and the scattering angle are shown in Fig. 1.

From expression (5) it can be seen that it is possible from measurements of the angular distribution and the polarization of the fluorescence to determine the alignment and polarization parameters of the atom, the knowledge of which makes it possible to form a judgment on the polarization of the nucleus.

EXPERIMENTAL SETUP

A diagram of the experimental setup for the measurement of the angular distribution and degree of polarization of the fluorescence radiation is shown in Fig. 2. A dye laser (rhodamine B) excited by a Q-switched solid-state laser based on an yttrium-aluminum garnet single crystal (LTIPCh-7) served as the source of the pulsed optical radiation. The light pulses had a duration of 10 ns, a pulse repetition frequency of from 12.5 to 100 Hz, and a power of up to 15 kW. Linear polarizers and quarter-wave plates placed along the radiation path were used to obtain light with linear or circular polarization.

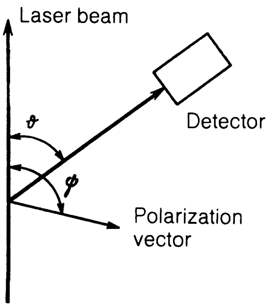

FIG. 1. Direction of scattered fluorescence radiation (ϑ) and its polarization vector (ψ).

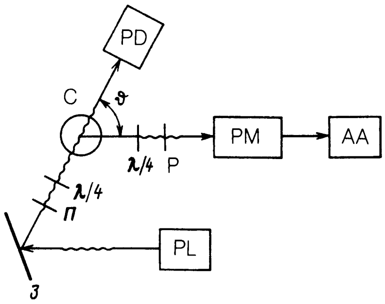

FIG. 2. Block diagram of the experimental setup: PL—pulsed dye laser, M—mirror, P—linear polarizer, $\lambda/4$—quarter-wave plate, C—sodium (^{23}Na) vapor cell, PD—photodioide for measuring the radiation power, PM—photomultiplier for counting photons, AA—pulse amplitude analyzer.

The resonance excitation of the Na atoms was carried out in a quartz cell with a volume ~ 1 cm^3. The cell could be heated to a temperature 200 °C using a coil through which an electric current was passed, and in this way it was possible to vary the sodium vapor pressure in the cell. To keep reabsorption of the photons emitted by the Na atoms to a minimum, their vapor pressure must be lower than 10^{10} at/cm^3, since the cross section of the interaction with the resonance is 10^{-10} cm^2. This corresponds to a sodium vapor pressure $4 \cdot 10^{-6}$ Torr in the cell.

The resonance-scattered fluorescence radiation was recorded with the help of a photomultiplier (FÉU-54) operating in the single-photon count regime. The amplitude propagation of the signals from the photomultiplier was measured by an AN-4096 pulse analyzer and served as a measure of the intensity of the resonance-scattered radiation at different angles and values of the polarization. The photomultiplier was rigidly mounted, and the direction of the laser beam into the cell was measured with the help of a system of mirrors, and in this way the angle ϑ was chosen at which the scattered radiation was measured. A photodiode served to monitor the intensity of the laser radiation passing through the cell.

To eliminate disorientation of the atoms by random fields, the cell was placed in a sufficiently strong field (up to 200 G), generated by a pair of Helmholtz coils and directed along the laser beam. To shield the photomultiplier from this field it was screened by a permalloy screen and situated 70 cm away from the Na atom excitation zone. The solid angle in which the scattered radiation was recorded was 0.05 sr.

As a control experiment to check that the setup was working correctly, we measured the degree of polarization and the angular distribution of fluorescence radiation during the irradiation of the Na vapor by linearly polarized light at the D 1 line. In this case the fluorescence radiation should be unpolarized and its angular distribution should be isotropic.[9] These measurements allowed us to introduce corrections for variation of the solid angle with variation of the angle ϑ and also for attenuation of the scattered radiation by the polaroids.

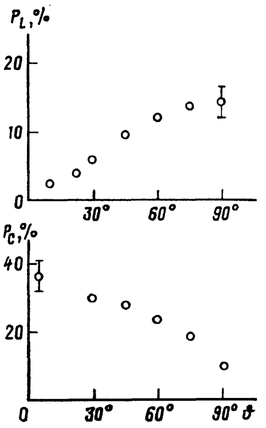

FIG. 3. Dependence of the degree of linear (P_L) and circular (P_C) polarization of the fluorescence on the scattering angle ϑ.

MEASUREMENT RESULTS

The described setup was used to measure the angular distribution and polarization of fluorescence radiation with the dye laser tuned to the D 1-line ($\lambda = 589.6$ nm) and the D 2-line ($\lambda = 589.0$ nm). The laser line width was 20 GHz, wherefore the excitation took place on both components of the hyperfine structure of the ground state $s_{1/2}$. The radiation power corresponded to the onset of saturation of the optical transition. The following experiments were carried out under these conditions.

1. By irradiating with laser light of one of the polarizations ($I'_\parallel$—the polarization vector lies in the scattering plane, or $I'_\perp$—perpendicular to this plane) we measured the dependence of the degree of linear polarization of the resonance fluorescence (P_L) on its angle of emission with respect to the laser beam (ϑ):

$$P_L(\vartheta) = \frac{I_\parallel(\vartheta) - I_\perp(\vartheta)}{I_\parallel(\vartheta) + I_\perp(\vartheta)}, \tag{6}$$

where $I_\parallel(\vartheta)$ and $I_\perp(\vartheta)$ correspond to the angles $\psi = 0$ and $\psi = \pi/2$ (Fig. 1). The degree of linear polarization is connected with the alignment parameter A_2^F by the relation

$$P_L(\vartheta) = \frac{3A_2^F \sin^2\vartheta}{4 + A_2^F(1 - 3\cos^2\vartheta)}. \tag{7}$$

2. The dependence of the relative variation of the intensity of the fluorescence (P'_L) on the angle ϑ was measured by varying the polarization of the laser radiation (from $I'_\parallel$ to $I'_\perp$):

$$P_L' = \frac{I_\perp' - I_\parallel'}{I_\perp' + I_\parallel'}, \tag{8}$$

$$P_L' = \frac{3A_2^F \sin^2\vartheta}{8 - A_2^F(2 + 3\sin^2\vartheta)}. \tag{9}$$

3. By irradiating with circularly polarized radiation we measured the degree of linear polarization at the angle 90°. It is connected with the alignment parameter by the relation

$$A_2^F = 4P_L/(3 - P_L). \tag{10}$$

4. Under the same conditions we measured the dependence of the degree of circular polarization P_C on the angle ϑ:

$$P_C(\vartheta) = \frac{I_+(\vartheta) - I_-(\vartheta)}{I_+(\vartheta) + I_-(\vartheta)}, \tag{11}$$

where $I_+(\vartheta)$ and $I_-(\vartheta)$ are the fluorescence intensities for circular polarization to the right and the left, respectively.

This quantity is determined by the alignment parameter as well as by the polarization:

$$P_C(\vartheta) = \frac{6P_F \cos\vartheta}{4 + A_2^F(1 - 3\cos\vartheta)}. \tag{12}$$

Examples of the measured distributions are shown in Fig. 3, and measured values of P_L and P_C as well as the parameters A_2^F and P_F obtained with the help of expressions (7) and (9) are given in Tables I and II.

We also measured the dependence of the degree of polarization of the fluorescence on the laser radiation power. One of these dependences (for the D 1-line with circular polarization) is shown in Fig. 4. For convenience of comparison with the calculations the degree of polarization of the nucleus (its determination is considered below) is the ordinate, and the parameter z, equal to the area of the laser pulses:

$$z = \int_0^\tau W(t)\,dt, \tag{13}$$

where $W(t)$ is the instantaneous power of the laser radiation and τ is the duration of the pulse, is the abscissa. If the pulse duration is less than the relaxation time of the excited level of the atom the degree of polarization does not depend on the pulse shape, and depends only on the area of the pulse.[10]

It is clear from Fig. 4 that for a certain power of the laser radiation there is observed a maximum of the degree of polarization of the nucleus. The magnitude of the power is determined by the Rabi oscillation frequency Ω:

$$\Omega = cWG(\lambda), \tag{14}$$

TABLE I. Alignment parameters obtained in the runs with linearly polarized radiation.

Transition	$P_L(90°)$	A_2^F	
		experiment	calculation
$s_{1/2} \rightarrow p_{1/2}$	0,14±0,02	0,19±0,03 * 0,16±0,03 **	0,20 0,20

*From the measurements by the direct approach (see Introduction).
**From the measurements by the indirect approach (see Introduction).

TABLE II. Alignment and polarization parameters obtained in the runs with circularly polarized radiation.

Transition	P_L (90°)	A_2^F		P_C (5°)	P_F	
		exp.	calc.		exp.	calc.
$s^1/_2 \to p^1/_2$	—	—	—	0,31±0,06	0,35±0,06	0,40
$s^1/_2 \to p^3/_2$	0,07±0,02	0,10±0,03	0,14	0,38±0,06	0,27±0,06	0,33

where $G(\lambda)$ is the transition matrix element and c is a proportionality coefficient. The maximum polarization corresponds to the power at which the Rabi oscillation period is close to the duration of the radiation pulse.

DISCUSSION OF RESULTS

From Tables I and II it can be seen that after one laser pulse (10^{-9} s) a significant degree of alignment of the atoms (up to 14%) as well as polarization (up to 35%) is attained. To assess the degree of orientation of the nuclei, it is necessary to compare the experimental values of the alignment and polarization parameters A_2^F and P_F with the calculated values, taking the hyperfine interaction into account. Towards this end we calculated the density matrix of the ensemble of atoms finding themselves in the excited state (the levels $p_{1/2}$ and $p_{3/2}$). The diagonal elements of the density matrix are proportional to the probabilities of the electric dipole transitions from the ground state to the excited state:

$$\rho_{JEM,\,J'F'M'} \propto W(JEM \to J'F'M'). \tag{15}$$

The transition probabilities can be expressed in terms of $3j$ and $6j$ coefficients:

$$W(JEM \to J'F'M') = c(2F+1)(2F'+1)\begin{pmatrix} F & 1 & F' \\ M & 1 & M' \end{pmatrix}\begin{Bmatrix} J & F' & I \\ F & J' & 1 \end{Bmatrix}. \tag{16}$$

The values of the parameters A_2^F and P_F calculated in this way are given in Tables I and II. Good agreement is seen between the experimental values and the calculated values which take the hyperfine interaction into account. This means, as was already noted above, that the angular momentum of a polarized photon absorbed by the atoms is distributed among the electron shell and the nucleus, as a consequence of which the nucleus becomes oriented. To describe the system of oriented nuclei one typically uses the alignment (A_2^I) and polarization (P_I) parameters. They are also expressed as in the case of atoms in terms of irreducible tensor operators:

$$A_2^I \equiv \overline{\langle T_{20}(I)\rangle} = \sum_{F,M} \frac{\langle FM|3I_z^2 - I^2|FM\rangle}{I(I+1)}, \tag{17}$$

$$P_I \equiv \overline{\langle T_{10}(I)\rangle} = \sum_{F,M} \frac{\langle FM|I_z|FM\rangle}{I}. \tag{18}$$

The parameters describing the system of oriented atoms and nuclei are connected by the relation

$$\langle T_{kq}(I)\rangle = \langle T_{kq}(F)\rangle \frac{\langle F\|T_k(I)\|F\rangle}{\langle F\|T_k(F)\|F\rangle} C_k^F, \tag{19}$$

where

$$C_1^F = \frac{F}{I}, \qquad C_2^F = \frac{F(2F+1)}{I(2I+1)},$$

$\|T_k(I)\|$ and $\|T_k(F)\|$ are the reduced matrix elements of the corresponding operator.

Making use of these relations, it is possible to obtain from the measured quantities A_2^F and P_F (Tables I and II) the values of the parameters A_2^I and P_I, which are given in Table III. From this table it can be seen that upon irradiation of the Na atoms by polarized laser radiation there arises a marked degree of orientation of the nuclei. It is maximum for the D 1-line since in this case all of the angular momentum transmitted to the atom is expended in changing the direction of its spin.

More detailed information on the mechanism of orientation of the nucleus can be obtained from the dependence of its degree of polarization on the laser radiation power. To calculate this dependence it is necessary to solve the evolution equation of the atomic density matrix.[11] Under the condition that the laser pulse duration is less than the fluorescence time and the hyperfine interaction in the ground state of the atom is much stronger than in the excited state, the expression for the degree of polarization of the nucleus has the form

$$P_I = \frac{100\%}{(2I+1)(2J+1)} \sum_{M=-F}^{I+J-1} \mathscr{P}_M(FIJJ')\sin^2\frac{\chi_M Z}{2}, \tag{20}$$

where $\mathscr{P}_M$ and χ_M are the polarization parameters. Expressions for $\mathscr{P}_M$ and χ_M and their concrete values for various values of I, J, and F were obtained in Ref. 11.

From expression (20) it follows that the dependence of

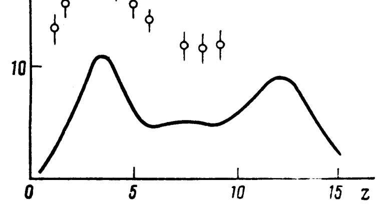

FIG. 4. Dependence of the degree of polarization of the ^{23}Na nucleus (P_I) on the parameter z [see Eq. (13)] during excitation by the D 1-line of circularly polarized radiation from one component of the hyperfine structure ($F = 1$ or 2): points—experiment, curve—theory [see Eq. (20)].

TABLE III. Alignment and polarization parameters of the nucleus.

Transition	Polarization	$A_2{}^I$	P_I
$s^1/_2 \rightarrow p^1/_2$	linear	0	0
$s^1/_2 \rightarrow p^1/_2$	circular	–	0,31
$s^1/_2 \rightarrow p^3/_2$	linear	0,09	0
$s_{1/2} \rightarrow p_{3/2}$	circular	0,11	0,20

the degree of polarization of the nucleus on the laser radiation power has an oscillatory character (see Fig. 4). The maximum value of P corresponds to the power at which there takes place only one induced transition to the excited state of the atom (the π-pulse regime) during the time of the radiation pulse.

Figure 4 compares experimental data for the polarization of the ^{23}Na nucleus as a function of the area of the laser pulse $z \sim W$ [see Eq. (13)] with the theoretical calculations [see Eq. (20)]. The theoretical calculations were carried out for the case in which the resonance excitation of the atomic transition is made up out of one component of the hyperfine splitting ($F = 1$ or 2). In actual fact the spectral width of the laser radiation contains both components of the hyperfine structure. In this case, strictly speaking, the two-level approximation no longer suffices. The polarization of the nucleus from the two components of the hyperfine structure was considered in Ref. 11 according to a three-level scheme. In this case the polarization of the nucleus depends on the phase shift between the modes, which correspond to resonance excitation of the two components of the hyperfine structure. If this shift varies stochastically during the time of the laser pulse, then the magnitude of the nuclear polarization will be the same as in the case of resonance excitation by monochromatic light from one component of the hyperfine structure. If the phase shift varies only little during the time of a pulse, then the magnitude of the polarization can be two times larger in comparison with the previous case.

Thus, from a comparison of theory and experiment it follows that

a) there exists qualitative agreement between the theoretical calculation, based on the assumption of stochastic phase shifts, and the experimental data;

b) the extent to which the polarization effect observed experimentally exceeds the theoretical value is apparently related to the fact that the phase shift between the two resonant modes is correlated over the entire extent of the laser pulse.

It stands to reason that a more detailed comparison of theory with experiment can be obtained in the case of resonance excitation by monochromatic laser radiation from each component of the hyperfine structure.

The above measurements of the degree of polarization of the ^{23}Na atom and of the ^{23}Na nucleus and their dependence on the laser radiation power show that at a radiation power density higher than 10^3 W/cm^2 Rabi oscillations of the population of the components of the hyperfine structure of the atomic levels arise. In this case a rapid polarization of the nucleus takes place after a time determined by the magnitude of the hyperfine splitting. This time can reach fractions of a nanosecond, which makes it possible to realize efficient polarization of short-lived nuclei.

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