there is a reduction also in the quantum efficiency for a single scattering of a photon, which—when allowance is made for α_g—is

$$\tilde{\omega}_0 = \frac{\tau_t}{\tau_{rad}} \frac{\alpha - \alpha_b}{\alpha}.$$

When $\tilde{\omega}_0$ is close to unity, the quantum efficiency decreases rapidly on reduction in $\tilde{\omega}_0$. Consequently, for $\tilde{\omega}_0(\omega_t) \approx 1$ the line width of the scattered radiation may be considerably less than Γ. It then follows from Figs. 4 and 5 that the degree of polarization of the radiation increases away from the line center, so that the average degree of polarization of a line may exceed greatly the degree of polarization at the center. In the case of strong inhomogeneous broadening $\Gamma' \gg \Gamma$ with a Lorentzian distribution of the frequency ω_t, Eqs. (55) and (56) should be modified by replacing Γ with Γ'.

We shall conclude by noting that the above theory applies also to the Rayleigh scattering of polarized light by defects in isotropic solid and liquid media, and—under certain conditions—it also describes the resonance scattering of acoustic phonons.[16,17]

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Phase relaxation investigation under conditions of appreciable spin polarization

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Measurements were made of the phase-memory time T_m of Yb^{3+} ions in $CaWO_4$ single crystals activated simultaneously with Yb^{3+} and Tb^{3+} ions, which have substantially different g-factors. Under conditions when T_m is determined by the dipole-dipole interactions between the Yb^{3+} and Tb^{3+} ions, a strong decrease of the relaxation rate T_m^{-1} is observed with decreasing temperature. This is due to the considerable polarization of the Tb^{3+} ions, for which the condition $kT < g\beta H$ is satisfied.

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1. INTRODUCTION

The overwhelming majority of EPR research has been performed under conditions when the high-temperature approximation is valid, i.e., $g\beta H \ll kT$ (the notation here is standard). Yet the courses of the spin-spin and spin-lattice relaxation should change substantially if appreciable spin polarization[1)] takes place, i.e., $g\beta H \gg kT$. Experiments when the low-temperature approximation is valid are of particular interest if the investigations are performed by the electron spin echo (ESE) method, which is highly effective in the study of relaxation processes. No such investigations were performed previously, apparently because of the need for obtaining very low temperatures: thus, at wavelengths ~3 cm we have $g\beta H \sim 0.5$ K so that the hard-

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to-get temperatures <0.3 K are needed.

We report here the results of an experimental investigation of relaxation processes by the ESE method under low-temperature conditions. We have used here certain features of the ESE method, which made it possible to satisfy the condition $g\beta H \gg kT$ at the usual helium temperatures in easily attainable magnetic fields. The phase-memory time T_m measured by the ESE method is determined in the general case by both spin-lattice and spin-spin interactions. The mechanisms that determine the observable rate of the phase relaxation (PR) have by now been investigated in sufficient detail.[1,2] It has been shown, in particular, that under conditions when the spin-lattice interactions are weak and exert no noticeable direct influence on the rate of the PR, so that T_m is determined by the dipole-dipole interactions, the major role is assumed by the mechanisms of instantaneous diffusion (ID) and spectral diffusion (SD).

The condition $kT < g\beta H$ is quite differently satisfied for ID and SD; to demonstrate this, we recall the gist of the ID and SD mechanisms. We shall refer to the spins that are excited by a microwave field of amplitude H_1 and produce the spin-echo signal as the spins A, while the remaining spins, which do not participate in the formation of the echo signals but can exert a substantial influence on the rate of the PR of spins A, will be designated spins B. If the ID process is effective, then the PR rate is determined by the fraction of spins A excited by the field H_1 and by the magnitude of their dipole-dipole interaction. In this case the aforementioned difficulty of satisfying the condition $kT < g\beta H$ is unavoidable. If the SD is the effective mechanism, then the PR rate of the spins A is determined by the dipole-dipole interactions of the spins A with the spins and by the rate W of reorientation of the spins B on account of the spin-lattice or spin-spin interactions. It turns out that in this case the condition $kT < g\beta H$ can be satisfied at easily obtainable magnetic fields and temperatures.

The idea of the experiment is the following. We choose for the investigation a sample containing two sorts of paramagnetic centers with substantially different g-factors and concentrations. This choice of the sample is governed by the following factors. First, if the concentration of the investigated spins (spins A) in the sample is negligible and the number of the other spins (spins B) is large, then the phase relaxation of spins A is determined mainly by the dipole-dipole interactions with spins B. Second, if the spins A are chosen to be paramagnetic centers with small g factor, so as to operate in a sufficiently strong (but easily attainable) magnetic field, while the spins B are centers with large g factors, then the condition $g\beta H > kT$ for the spins B will be satisfied at easily attainable magnetic fields and temperatures. It is precisely under these conditions that we have investigated the PR in the present study.

TABLE I. Experimental data on the phase relaxation of Yb^{3+} ions in $CaWO_4 + Yb^{3+} + Tb^{3+}$ single crystals with orientation $\theta = 0°$.

№	Sample	T, K	Dependence of T_m on the amplitude of H_1	$\varkappa$	T_m, μsec
I	$CaWO_4$ + 0.01% Yb^{3+} + 0.1% Tb^{3+}	4.23	None	3/2	8.5
		1.64	Weak	≥3/2	35–40
II	$CaWO_4$ + 0.01% Yb^{3+} + 1% Tb^{3+}	4.23	None	3/2	1.2
		1.6		3/2	4.6
III	$CaWO_4$ + 0.1% Yb^{3+} + 0.1% Tb^{3+}	4.24	Strong	≤3/2	6–7.5
		1.53	Strong	1–3/2	13–30
IV	$CaWO_4$ + 0.002% Yb^{3+} + 0.1% Tb^{3+}	4.2	None	≤2	14
		1.6	None	2	65

2. EXPERIMENTAL RESULTS

As the investigation object we have chosen a $CaWO_4$ single crystal simultaneously activated by Yb^{3+} and Tb^{3+} ions. The g-factors of the impurity ions in the tetragonal field of the $CaWO_4$ crystal are strongly anisotropic and their values are $Yb^{3+} - g_{\parallel} = 1.054, g_{\perp} = 3.014$,[3] and $Tb^{3+} - g_{\parallel} = 17.777, g_{\perp} < 0.15$.[4] We have investigated four $CaWO_4$ samples with different concentrations of the Yb^{3+} and Tb^{3+} ions, namely: $CaWO_4$ + 0.01% Yb^{3+} + .01% Tb^{3+} (sample I); $CaWO_4$ + 0.01% Yb^{3+} + 1% Tb^{3+} (sample II); $CaWO_4$ + 0.1% Yb^{3+} + 0.01% Tb^{3+} (sample III); $CaWO_4$ + 0.002% Yb^{3+} + 0.1% Tb^{3+} (sample IV). The role of the spins A was assumed in this case by the Yb^{3+} ions, and that of spins B by the Tb^{3+} ions. The phase-memory times were measured at a frequency ~9.4 GHz with a spin-echo spectrometer similar to that described earlier.[5]

The experimentally observed decrease of the spin-echo signal amplitude with increasing time τ between the microwave pulses that form the echo signal can always be well described by a relation in the form

$$A(2\tau) = \exp\{-(2\tau/T_m)^{\varkappa}\}, \qquad (1)$$

where $\varkappa$ takes on values from 1 to 2.

It follows from the data on the g-factor that if T_m is measured with Yb^{3+} ions in the orientation $\theta = 0°$ (θ is the angle between the magnetic field and the c axis of the crystal), i.e., working in a 6.23 kG magnetic field, we get for the Tb^{3+} ions $g\beta H = 7.6$ K. At the lowest temperature obtained in our experiments, namely ~1.5 K, the polarization of the Tb^{3+} ions in this orientation is

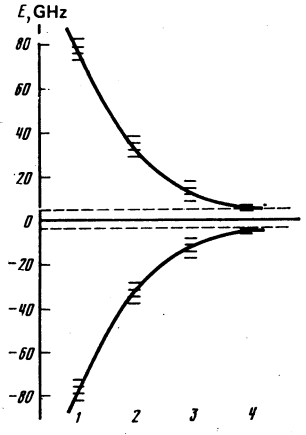

FIG. 1. Energy level scheme of Tb^{3+} ion in $CaWO_4$ crystals at certain values of the angle θ in a magnetic field corresponding to the resonant frequency of the Yb^{3+} ion. 1) $\theta = 0°$, $H_{res} = 6.23$ kG; 2) $\theta = 30°$, $H_{res} = 3.1$ kG; 3) $\theta = 60°$, $H_{res} = 1.95$ kG; 4) $\theta = 90°$, $H_{res} = 1.71$ kG. Dashed lines—Zeeman splitting of the Yb^{3+} ion.

very substantial, $p = 0.987$. On deviating from the orientation $\theta = 0°$, i.e., when the EPR signal from Yb^{3+} is observed in a weaker magnetic field and the g-factor of Tb^{3+} decreases, the values of $g\beta H$ and of the Tb^{3+} polarization decrease. Thus, at $\theta = 30°$ we have $g\beta H = 3.2$ K and $p = 0.789$ (at $T \sim 1.5$ K). At $\theta = 60°$ and more so at $\theta = 90°$ the value of $g\beta H$ for the Tb^{3+} ions in the resonant magnetic field of the Yb^{3+} ions becomes even smaller, and the polarization is insignificant (even at $T \sim 1.5$ K).

Since we are primarily interested in data on the phase-memory times of the Yb^{3+} ions under conditions of appreciable spin polarization of the Tb^{3+} ions, we present in the table some experimental results obtained at low temperature at the orientation $\mathbf{H} \| c$ ($\theta = 0°$). It is seen from the table that the most suitable object for our problem is sample IV, in which no contribution whatever is made to the phase relaxation by the instantaneous spectral diffusion, as follows from the independence of T_m of the amplitude H_1 (see Fig. 2), and the times T_m are long enough to permit expansion of the range of investigated relaxation times.

The measured phase-memory times T_m of the Yb^{3+} ions in sample IV are shown in Fig. 3 for various orientations of the magnetic field relative to the c axis of the crystal. The figure shows also data from Refs. 6 and 7 on the spin-lattice relaxation times T_1 of the ions Yb^{3+} and Tb^{3+} in $CaWO_4$. It is seen from Fig. 3 that at $T > 6$ K the times T_m become much shorter, a fact that can be attributed to the substantial role of the time of the spin-lattice relaxation of the Yb^{3+} and Tb^{3+} ions.

We consider now the measured T_m of the Yb^{3+} ions in the range 1.5–6 K. As a rule, at these temperatures the times T_m are independent of temperature (see, e.g., the preceding study[8] of T_m for Yb^{3+} in $CaWO_4$ in the absence of the Tb^{3+} impurity), but is must be noted that the published data were obtained under conditions of negligible spin polarization. The results of the measurements of T_m of the Yb^{3+} ions in $CaWO_4 + Yb + Tb$ in the 1.5–6 K range can be described as follows: With decreasing temperature, at $\theta = 0°$, when the polarization increases considerably the time T_m also increases strongly; at $\theta = 30°$, when the polarization is weaker, the temperature dependence of T_m is weaker; at $\theta = 60°$ and $\theta = 90°$, when the polarization is negligible, there is practically no temperature dependence of T_m.

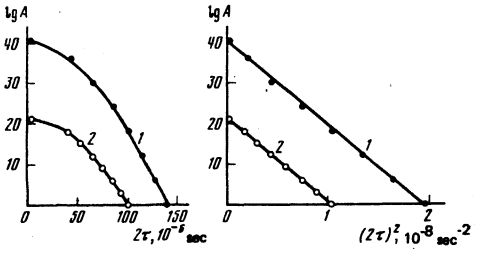

FIG. 2. Dependence of the amplitude of the spin echo of the Yb^{3+} ion on the interval τ between the pulses in the sample $CaWO_4 + 0.002\%$ $Yb^{3+} + 0.1\%$ Tb^{3+} at various microwave power levels ($\theta = 0°$, $T \approx 1.6$ K): 1) $P = 0$ dB, 2) $P = 11$ dB.

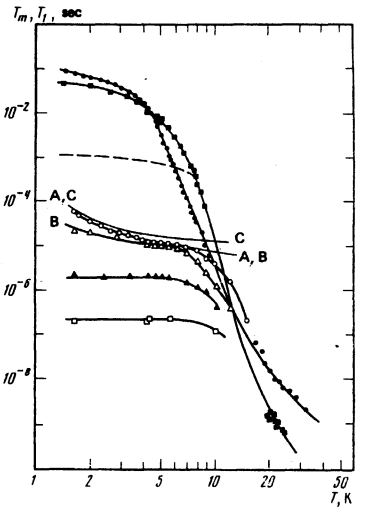

FIG. 3. Temperature dependence of the times T_m of the Yb^{3+} ions for sample IV and of the spin-lattice relaxation time T_1 of the ions Yb^{3+} and Tb^{3+}. ○—T_m, $\theta = 0°$; △—T_m, $\theta = 30°$; ▲—T_m, $\theta = 60°$; □—T_m, $\theta = 90°$; ●—$T_1(Yb^{3+})$, $\nu \approx 9.4$ GHz; ■—$T_1(Tb^{3+})$, $\nu \approx 36.0$ GHz. Dashed line—times T_1 of Tb^{3+} ions, calculated from formula (13). Lines A—A and B—B are the functions $f_1(T)$ respectively for $g\beta H = 7.6$ K and 3.2 K. Line C—C is the function $f_2(T)$ for $g\beta H = 7.6$ K.

We note also that in the same temperature range we have $\varkappa = 2$ for $\theta = 0°$ and $\theta = 30°$ and $\varkappa \approx 1$ for $\theta = 60°$ and $\theta = 90°$. Measurements in $CaWO_4$ crystals containing only Yb^{3+} ions or only Tb^{3+} ions have shown that at $T < 6$ K the times T_m are independent of temperature and are equal (at $\theta = 0°$) to $\sim 6 \times 10^{-4}$ sec for 0.022% Yb^{3+} and $\sim 2 \times 10^{-7}$ sec for 0.1% Tb^{3+}.

3. DISCUSSION OF RESULTS

In our case, i.e., when spins of a second sort (Tb^{3+} ions) are present in the sample in large amounts, the times T_m of the Yb^{3+} ions are determined mainly (and in the case of sample IV, completely) by the spectral diffusion due to the dipole-dipole interaction with the Tb^{3+} spins. Models of the spectral diffusion were thoroughly discussed in the literature (see, e.g., Refs. 1, 2, and 9). The kinetics of the decay of the spin-echo amplitude and of the rate of phase relaxation for two model processes, in the approximations of slow ($W\tau \ll 1$) and fast ($W\tau \gg 1$) spectral diffusion, are determined by formula (1) and are characterized by the following values of the parameters $\varkappa$ and T_m.

For the "fast jumps" model[9]

$$\varkappa = 2,\ T_m^{-1} = 0.7\Delta\omega_{1/2}^{1/2}W^{1/2} \text{ at } W\tau \ll 1, \tag{2}$$

$$\varkappa = {}^3/_2,\ T_m^{-1} = 0.6\Delta\omega_{1/2}^{2}W^{-1} \text{ at } W\tau \gg 1. \tag{3}$$

For the Gauss-Markov model[2]

$$\varkappa = {}^3/_2,\ T_m^{-1} = 0.5\Delta\omega_{1/2}^{3/2}W^{1/2} \text{ at } W\tau \ll 1, \tag{4}$$

$$\varkappa = {}^1/_2,\ T_m^{-1} = 0.9\Delta\omega_{1/2}^{2}W^{-1} \text{ at } W\tau \gg 1. \tag{5}$$

By $\Delta\omega_{1/2}$ we denote the shift of the frequency of spin A following a change of the local magnetic field at the location of the spin A on account of reorientation of the spins B. In the high-temperature approximation, $\Delta\omega_{1/2}$

is the dipole-dipole width of the EPR line and is given by[2]

$$\Delta\omega_{1/2}=\frac{4\pi^2}{3^{3/2}}\frac{g_A g_B\beta^2}{\hbar}n_B =2.07\cdot10^{-13}\,g_A g_B n_B, \tag{6}$$

where n_B is the number of spins B per cm^3.

The experimental results for sample IV show that at appreciable spin polarization, i.e., at $\theta=0°$ and $\theta=30°$, the kinetics of the decay of the echo signal is described by expression (2), which corresponds to the model of fast jumps.

Since the amplitude of the frequency shift $\Delta\omega_{1/2}$ does not depend on the population difference of the spin levels, the temperature dependence of the phase relaxation of the spins A should be determined only by the rate W of the flips of the spins B.

Hoping that the experimental results can identify the factor governing the value of W, either spin-lattice relaxation of the spins B (designated W_1) or flip-flop (FF) processes (designated W_2), we shall attempt to determine the temperature dependences of these quantities.

We denote by W_+ the probability of finding the spin on the lower level, by W_0 the probability of finding the spin on the upper levels, by W_{+-} and W_{-+} the probabilities of spin transitions from the lower to the upper levels and vice versa, and put $T_1^{-1}=W_{+-}+W_{-+}$. It is obvious that

$$W_1=W_+w_{+-}+W_-w_{-+}. \tag{7}$$

Recognizing that

$$\frac{W_+}{W_-}=\frac{w_{-+}}{w_{+-}}=\frac{1+p}{1-p}$$

we easily obtain

$$W_1=\tfrac{1}{2}(1-p^2)T_1^{-1}=(1-p^2)W_1^0. \tag{8}$$

Using (8), we write down the expressions for the amplitude of the spin echo (1) and for the rate of phase relaxation (3) with allowance for the spin polarization for the fast-jumps model, under the condition that W is determined by the spin-lattice relaxation:

$$A(2\tau)=\exp[-(1-p^2)T_1^{-1}\Delta\omega_{1/2}\tau^2], \tag{9}$$

$$T_m^{-1}=0.5\,\Delta\omega_{1/2}(1-p^2)^{1/2}(T_1^{-1})^{1/2}. \tag{10}$$

In the case of FF processes, the rate of flipping of the spins B can be easily shown to be again proportional to $1-p^2$, so that we can write

$$W_2=(1-p^2)W_2^0. \tag{11}$$

Thus, an appreciable spin polarization produces in the expressions for W_1 and W_2 the same factor $1-p^2$. This does not mean, however, that the character of the temperature dependence is the same for W_1 and W_2, since the change of temperature does not affect W_2^0, and

$$W_1^0=\tfrac{1}{2}T_1^{-1}\sim\mathrm{cth}(g\beta H/2kT)=1/p.$$

Consequently in the fast-jumps model the temperature dependence of the rate of the phase relaxation (2) is determined by the factor $f_1(T)=[(1-p^2)/p]^{1/2}$ if the principal role is played by spin-phonon interactions, and by the factor $f_2(T)=(1-p^2)^{1/2}$ when the influence of the FF processes predominates. In practice, however, when the polarization is appreciable and approaches its maximum value $p=1$, the difference between the functions f_1 and f_2 vanishes.[3)] Under these conditions it is possible to make an unequivocal choice between the two discussed causes of the flipping of the spins B on the basis of the available experimental results only by a comparison with the data on the absolute values of W_1^0 and W_2^0.

We turn now to the experimental results for sample IV (see Fig. 3), for which the PR rate is completely determined by the SD mechanism. We confine ourselves first to $T<6$ K and to orientations $\theta=0°$ and $\theta=30°$, inasmuch as in this case the condition of high spin polarization is satisfied. It is seen from Fig. 3 that the change of the rate of the PR with changing temperature is well described by the function $f_1(T)$ and somewhat worse by the function $f_2(T)$. The values of W_1^0 and W_2^0 can be estimated in the following manner. The times of the SLR of the Tb^{3+} ions in $CaWO_4$ were measured earlier[7] at 36 GHz and it turned out that $T_1^{-1}=10.4\times\mathrm{coth}(1.73/2T)$ sec^{-1}. The SLR theory for non-Kramers ions[11] yields the following dependence of the SLR rate on the intensity of the applied static magnetic field:

$$T_1^{-1}\sim H^3\,\mathrm{cth}(g\beta H/2kT), \tag{12}$$

we can therefore write for our crystal (at $\theta=0°$ and $\Delta=7.6$ K)

$$T_1^{-1}=876\,\mathrm{cth}\left(\frac{7.6}{2T}\right)=876\frac{1}{p}. \tag{13}$$

The relaxation times calculated from (13) are also shown in Fig. 3.

To make use of Eq. (2) we must know $\Delta\omega_{1/2}$. From (6), assuming $n_B=1.26\times10^{19}$ ions/cm^3 for $\theta=0°$, we have $\Delta\omega_{1/2}=4.9\times10^7$ sec^{-1}. For the temperature $T=3.8$ K and the orientation $\theta=0°$ we get $T_m=1.3\times10^{-5}$ sec, very close to the experimental 1.6×10^{-5} sec.

An analgous calculation for $\theta=30°$ yields $T_1^{-1}=65.35/p$, $\Delta\omega_{1/2}=8.67\times10^7$ sec^{-1} ($g_A=2.16$ and $g_B=15.395$). At $T=3.2$ K we obtain $T_m(\mathrm{calc.})=2.06\times10^{-5}$ sec, which is again close to the experimental (see Fig. 3) $T_m(\mathrm{exp.})\sim1.8\times10^{-5}$ sec.

Thus, the experimental results on the time T_m of the Yb^{3+} ions in the orientations $\theta=0°$ and $\theta=30°$ are very well described by the theory both qualitatively and quantities, if it is assumed that the PR is determined by the SD, and W is determined by the rate of the reorientation of the Tb^{3+} on account of their spin-lattice relaxation. The function $f_2(T)$ (line B-B on Fig. 3) describes the experimental results less accurately than $f_1(T)$, thus indicating a negligible contribution of FF processes on the PR. Unfortunately, we are not in a state to calculate W_2^0 accurately, since there is no theoretical analysis of FF processes in a system of non-Kramers rare-earth ions. We shall therefore estimate of this quantity. According to Ref. 2

$$W_2^0\approx2\pi\frac{(\Delta\omega_{dip})^2}{\Delta\omega_{ex}}, \tag{14}$$

where $\Delta\omega_{dip}$ is the homogeneous part of the resonance-line width and is due to the dipole interactions, while $\Delta\omega_{st}$ is the inhomogeneous EPR line width due mainly to all possible distortions of the crystal field. If the g-factor were isotropic, we would have

$$\Delta\omega_{dip}\sim g^2\beta^2\hbar^{-1}n_B. \tag{15}$$

However, the behavior of a non-Kramers ion is described by the spin Hamiltonian[4]

$$\mathcal{H}=\Delta_x S_x+\Delta_y S_y+g_\parallel\beta HS_z\cos\theta, \tag{16}$$

in which $g_\perp=0$, and consequently the probability of the FF processes differs from zero only on account of the first two terms of the Hamiltonian, which determines the initial splitting $\Delta_0=(\Delta_x^2+\Delta_y^2)^{1/2}$. Therefore, when $g\beta H\cos\theta>\Delta_0$, formula (15) can be used to estimate $\Delta\omega_{dip}$ if we put

$$g\beta=\frac{\Delta_0}{\sqrt{2}H\cos\theta}=\frac{g_\parallel\beta}{\sqrt{2}}\left(\frac{\Delta_0}{\Delta}\right).$$

We have thus

$$W_2^0\sim\frac{\pi g_\parallel^4\beta^4}{2\hbar^2}\left(\frac{\Delta_0}{\Delta}\right)^4\frac{n_B^2}{\Delta\omega_{st}}. \tag{17}$$

To estimate $\Delta\omega_{st}$, measurements were made of the EPR of the Tb^{3+} ions. These measurements have shown that on going from the 3-cm to the 8-mm band the resonance-line width increases from ~3 G to ~30 G at the orientation $\theta=0°$. Extrapolation to a magnetic field $H\sim6.2$ kG yields $\Delta H_{st}=100$ G and $\Delta\omega_{st}\sim2\pi\times10^{10}$ sec^{-1}. We have multiplied here the width of the individual line by $2I+1=4$, to take into account the effect of the hyperfine structure. Using the values presented above for the quantities in (17), we obtain $W_2^0\sim30$ sec^{-1}. Since $W_1^0\sim10^3$ and 10^2 sec^{-1} for the cases $\theta=0°$ and $\theta=30°$, respectively, our conclusion that the influence of the FF processes can be neglected compared with the role of the spin-lattice relaxation in the case of appreciable polarization becomes fully convincing.

We have considered so far the results obtained at $T<6$ K for $\theta=0°$ and $\theta=30°$. At $\theta=60°$ and $\theta=90°$ the times T_m of the Yb^{3+} ions become much shorter (see Fig. 3) and do not depend on the temperature. Since W_2^0 increases with increasing θ and W_1^0, on the contrary, decreases with increasing θ, it can be assumed that at $\theta=60°$ and $\theta=90°$ the decisive role in the SD mechanism is assumed by the FF processes of the Tb^{3+} ions. Favoring this assumption are both the independence of T_m of the temperature and the estimates of the values of W (at $\theta=60°$ we have $W_1^0\sim10^1$ sec^{-1} and $W_2^0\sim10^4$ sec^{-1}).

Finally, measurements of the PR times directly on the Tb^{3+} ions (spins B) has made it possible to determine experimentally that for $\Delta\sim0.5$ K the rate of the PR decreases with increasing θ and $W_2^0\sim10^6$ sec^{-1}. These results are in good agreement with estimates based on formulas (1) and (17).

As for the results obtained for T_m at $T>6$ K, their strong temperature dependence leads to the conclusion that a substantial contribution to the observed rate of the PR of the Yb^{3+} ions is made by the spin-lattice relaxation processes of the Yb^{3+} ions themselves as well as by that of the Tb^{3+} ions. A noteworthy still unexplained fact is that at $T>6$ K an abrupt shortening of the times T_m is observed when the orientation is changed from $\theta=0°$ to $\theta=90°$ (see Fig. 3).

We shall discuss also some experimental results obtained with other samples (see the Table). In sample III, which has a larger concentration of Yb^{3+} ions than samples I, II, and IV, the observed dependence of T_m on H_1 is quite strong. This points to a substantial role played in this sample by the instantaneous-diffusion mechanism, and hence to a weakening of the temperature dependence of T_m, which is connected with the polarization of the Tb^{3+} ions. In sample I the times T_m are close to those of sample IV, and the difference is apparently due to the fact that the actual concentration of the Tb^{3+} ions is somewhat higher in sample I than in sample IV. In sample II, just as in sample IV, the rate of the PR is determined entirely by the SD mechanism. It must be noted that the tenfold increase of the Tb^{3+} ion concentration in sample II relative to IV produced a more than tenfold shortening of T_m. This cannot be explained if it is assumed that W is determined by the value of W_1, for in this case T_m should be changed by a factor $\sqrt{10}\sim3$ on account of the change of $\Delta\omega_{1/2}$. It can be assumed that in sample II, at all θ, the time T_m is completely determined by the FF processes of the Tb^{3+} ions. This assumption is also corroborated by the fact that at $\theta=0°$ the function $f_2(T)$ agrees well with the experimental results.

4. CONCLUSION

The success of the experimental investigation of the phase relaxation under conditions of considerable spin polarization was due to the application of the electron spin echo procedure to a system containing spins of two kinds with substantially different g-factors (one must actually have $g_B/g_A>10$) and different concentrations (actually $n_A\leq0.01\%$ and $n_B\sim0.1\%$). In addition it is necessary to have a sufficiently large range (variation by a factor 5 or 6) of temperatures, in which the time T_m is determined only by the dipole-dipole interactions of the investigated electron spins. It is therefore necessary to choose a crystal containing a small number of nuclear spins, or else the nuclear magnetic moments must be small. Of course, some of these conditions will be determined in each concrete case by the technical means at the experimenter's disposal (working frequency, ranges of magnetic fields and temperatures).

Compared with the usual pulse procedures used to measure the times of the spin-lattice relaxation, an attractive feature of the method described here is the possibility of studying, in a wide range, the dependence of the relaxation on the intensity of the static magnetic field and on its orientation.

In the case investigated by us, the spin-lattice relaxation turned out to be more effective than the FF processes under conditions of high spin polarization, this being due to the use of a system B consisting of rare-earth ions with an even number of electrons. It is undoubtedly of interest to carry out similar experiments

on a sample that does not contain B particles in the form of Kramers ions.

Although we have considered here mainly experimental results obtained under conditions of high spin polarization, it is nevertheless of definite interest to interpret the remaining experimental results (the values of T_m and the kinetics of the echo decay in all the samples in a wide range of temperatures and at all orientations). This calls for additional experimental and theoretical research.

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[1)] By polarization, as usual, is meant the quantity $p = (n_+ - n_-)/(n_+ + n_-) \equiv \tanh(g\beta H/2kT)$, where n_+ and n_- are the numbers of the paramagnetic centers whose spins are oriented along and against the magnetic field, respectively.

[2)] The ion Tb^{3+} is a non-Kramers ion ($S = \frac{1}{2}, I = \frac{3}{2}$) having an initial splitting $\Delta_0 = 8.131$ GHz and a hyperfine-structure constant $A = 252.5$ G $\equiv 6.284$ GHz.[4] Figure 1 shows the positions of the energy levels of the Tb^{3+} ion in $CaWO_4$ for certain values of the angles θ. The interval between the levels with $m = 0$ is determined by the expression $\Delta = [(g_{\parallel}\beta H\cos\theta + Am)^2 + \Delta_0^2]^{1/2}$. By the quantity $g\beta H$ for the Tb^{3+} ion is meant here throughout the value of Δ under the assumption $A = 0$.

[3)] The authors thank V. A. Atsarkin for calling their attention for the error in the interpretation of our experimental results[10] through neglect of this circumstance.

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ERRATUM

Erratum: Narrow nonlinear nonresonances in a three-level system [Sov. Phys. JETP 51, 851–855 (1980)]

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In Appendix I, on page 854, the following expression was left out after equation (I.3):

$$D^{\alpha} = \delta_{21}{}^{\alpha}\delta_{31}{}^{\alpha}\delta_{23}{}^{\alpha} - \delta_{31}{}^{\alpha}\hbar^{-2}|V_{31}{}^{\alpha}|^2 - \delta_{23}{}^{\alpha}\hbar^{-2}|V_{23}{}^{\alpha}|^2$$

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