Alteration of the magnetization of a paramagnetic crystal by a light pulse

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We consider the possibility of noninertial change of the magnetization of a crystal by a light pulse as a result of adiabatic Stark inversion of the Zeeman sublevel on the leading front of the pulse. A light-pulse model that admits of an exact solution of the Schrödinger equation is constructed and yields an expression for the crystal-magnetization time dependence that is determined by characteristic switch-on time of the field. Crystals with triplet and quartet ground states of the paramagnetic impurities are considered by way of example. The role of inhomogeneous broadening is analyzed.

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1. INTRODUCTION

Propagation of electromagnetic radiation through a matter magnetizes the latter. For optical frequencies this phenomenon was first considered by Pershan[1] and called the inverse Faraday effect (IFE). It was shown[1] from general thermodynamic considerations that for a nonmagnetic substance the magnetization is proportional to the degree of circular polarization of the radiation.

In Ref. 2 was developed a nonstationary microscopic theory of the IFE for paramagnetic crystal, with account taken also of the reaction of the medium on the electromagnetic wave—the nonlinear change of the Stokes parameters of the radiation by the propagation in the medium. In particular, it was shown that in the presence of a constant magnetic field **B** the change of the crystal magnetization is connected not only with the degree of circular polarization of the radiation but also with the total intensity I: in a magnetoactive medium the magnetization changes in proportion to BI. In the nonstationary regime, in addition, a noninertial change of the magnetization is possible on account of stimulated Raman scattering (SRS) of the light pulse by the Zeeman sublevels of the paramagnetic ion, which are split by the field **B** and have nonequilibrium populations at sufficiently low temperatures. The stationary Pershan mechanism, on the other hand corresponds from the microscopic viewpoint to a relatively slow change, connected with the relaxation processes, of the population of the Zeeman sublevels, the distances between which are altered by the electromagnetic radiation on account of the quadratic Stark effect.

For the IEF to set in as a result of Raman scattering of light it is necessary that the width of the radiation spectrum be large enough, not less than the Zeeman splitting in the field **B**. Obviously, the Raman scattering is stimulated only in this case.

We consider in this paper one other possibility of noninertial alteration of the crystal magnetization, for which no broad-spectrum radiation is needed. We have in mind adiabatic Stark inversion of the Zeeman sublevels on the leading front of the light pulse. The physical picture is illustrated by Fig. 1. Assume no radiation prior to the instant t_1 and that the splitting of the pair of Zeeman sublevel is equal to δ. At $t > t_1$ the distance between levels changes because of the Stark effect, and under certain conditions quasicrossing of the levels is possible at a certain instant t_2. Obviously, if the time $t_2 - t_1$ is short compared with the relaxation times, but long enough to exclude nonadiabatic transitions between levels in the quasicrossing region, population inversion takes place at $t > t_2$ and leads to a change of the crystal magnetization.

In a number of recent papers, the effect of adiabatic inversion was considered under conditions of one-photon[4] and three-photon[5] resonances. At two-photon resonance, however, a particular case of which is the IFE mechanism considered here, the problem no longer contains a small parameter (see Ref. 6) and the theory turns out to be more complicated. To consider adiabatic inversion in two-photon resonance it is necessary to solve the problem exactly. Such a solution was obtained in Ref. 6 for several model dependences of the field intensity on the time. The results of Ref. 6 are used here to consider the proposed mechanism of noninertial reversal of crystal magnetization by light.

Section 2 contains a derivation of the general equations, while Secs. 3 and 4 deal with their application to triplet and quartet states. The role of inhomogeneous broadening of the states of the impurity ions is the subject of Sec. 5, which contains also numerical estimates of the effect. A brief exposition of the present results was given in Ref. 7.

2. GENERAL EQUATIONS

Let a plane quasimonochromatic optical wave of frequency ω propagate in a cubic axis along one of the axes. A constant magnetic field **B** is applied in the same direction. It is assumed that the rise time of the optical pulse, defined as

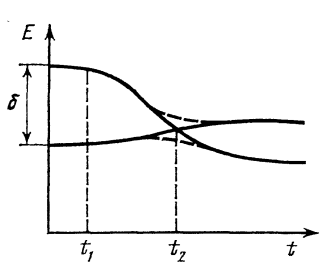

FIG. 1.

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the time interval it takes to reach a stationary or maximum value after it is turned on, is much shorter than all the relaxation times in the impurity-ion system. We assume also that only the ground state of the ions is populated.

Under these assumptions, the behavior of the triplet and quartet states of the ions in the field can be described by an effective two-level system. The triplet state is characterized by an effective spin $S=1$, and in the assumed geometry the field mixes the sublevels $m=1$ and $m=-1$. The sublevel $m=0$ is not magnetoactive, and will therefore not be considered.

The quartet states have an effective spin $S=3/2$ and the field mixes the sublevels of two separate pairs: $m=-3/2$, $1/2$ and $m=-1/2$, $3/2$. The explicit form of the effective Hamiltonian of the interaction of the radiation with the paramagnetic ions is given for the general case in Refs. 2 and 3. The actual Hamiltonian that determines the Stark shifts of the triplet sublevels is considered in Sec. 3 of the present paper.

Writing the density matrix of the two-level system in the form

$$\rho=\frac{1}{2}\begin{pmatrix}1-w & u+iv\\ u-iv & 1+w\end{pmatrix}, \tag{1}$$

we can obtain the kinetic equations for the quantities u, v, and w (Ref. 2):

$$\begin{aligned}\partial u/\partial t&=\varkappa\xi_1 Iw-[\delta+(\alpha+\beta\xi_2)I]v,\\ \partial v/\partial t&=[\delta+(\alpha+\beta\xi_2)I]u-\lambda\xi_3 Iw,\\ \partial w/\partial t&=(\lambda\xi_3 v-\varkappa\xi_1 u)I.\end{aligned} \tag{2}$$

Here I is the radiation intensity and ξ_j are the Stokes parameters. The radiation is assumed to be completely polarized, so that $\xi_1^2+\xi_2^2+\xi_3^2=1$. The quantity δ determines the Zeeman splitting of the sublevels in the absence of radiation:

$$\delta=(\mu_m-\mu_n)\mathbf{B}, \tag{3}$$

where μ_m and μ_n are the magnetic moments of the sublevels. The parameters α, β, $\varkappa$, and λ are determined by the matrix elements of the Hamiltonian of the interaction of the ion with the radiation[2]:

$$\begin{aligned}\alpha&=(1/4\sqrt{3})\,[\langle n|T_{\Gamma_3\theta}|n\rangle-\langle m|T_{\Gamma_3\theta}|m\rangle],\\ \beta&=1/4[\langle m|T_{\Gamma_4 0}|m\rangle-\langle n|T_{\Gamma_4 0}|n\rangle],\\ \varkappa&=(1/2i)\langle m|T_{\Gamma_5 0}|n\rangle,\quad \lambda=1/2\langle m|T_{\Gamma_3\varepsilon}|n\rangle,\\ T_{\Gamma_3\theta}&=(1/2\sqrt{3})\,(2D_{zz}^{(+)}-D_{xx}^{(+)}-D_{yy}^{(+)}),\\ T_{\Gamma_3\varepsilon}&=1/2\,(D_{xx}^{(+)}-D_{yy}^{(+)}),\quad T_{\Gamma_4 0}=iD_{xy}^{(-)},\quad T_{\Gamma_5 0}=D_{xy}^{(+)},\\ D_{ij}^{(\pm)}&=1/2\{d_i(G^{(-)}\pm G^{(+)})d_j\pm d_j(G^{(-)}\pm G^{(+)})d_i\},\\ G^{(\pm)}&=\sum_\lambda\frac{|\lambda\rangle\langle\lambda|}{\omega_{\lambda 0}\pm\omega},\end{aligned} \tag{4}$$

$\mathbf{d}$ is the electric-dipole-moment operator, and the summation is over all the excited states $|\lambda\rangle$ of the ion with energies $\omega_{\lambda 0}$ reckoned from the ground-state energy. The quantum numbers m and n denote the eigenvalues of the effective spin of the ground state: $m=-n=1$ for the triplet; $m=3/2$, $n=-1/2$ or $m=1/2$. $n=-3/2$ for the quartet. In accordance with the selection rules (see Ref. 2), $\alpha=0$ for the triplet states. For the quartet states the parameter α for the pairs $(-3/2,\ 1/2)$ and $(-1/2,\ 3/2)$ has opposite signs.

For Kramers doublets, the magnetic sublevels are not intermixed by the radiation at the magnetic field orientation and at the radiation propagation direction considered here. In Eqs. (2) we have in this case $\varkappa=\lambda=0$ (see Ref. 2), and the population difference is not altered by the optical radiation. For this reason the impurity ions whose ground state is a Kramers doublet are not considered in the present paper.

The initial conditions for Eqs. (2), under the condition that the ion-sublevel splitting in external fields is much smaller than the crystal temperature in energy units Θ, are of the form

$$u(-\infty)=v(-\infty)=0,\qquad w(-\infty)=\delta/Q\Theta, \tag{5}$$

where $Q=3$ for triplets and $Q=4$ for quartets.

The crystal magnetization can be expressed in this case in terms of the difference w of the sublevel populations:

$$M_x=M_y=0,\qquad M_z=N(\mu_{mz}-\mu_{nz})w, \tag{6}$$

where N is the density of the paramagentic ions in the case when the ground state of the ions is the quartet $w=w_{1/2}+w_{3/2}$, where $w_{1/2}$ and $w_{3/2}$ are the populations of the sublevels with projections $m=1/2$ and $3/2$.

We assume below that the density of the paramagnetic ions or the thickness of the crystal is small enough to neglect the change of the radiation polarization in the course of propagation. An estimate of the conditions needed for this purpose is given in Ref. 2.

The initial conditions (5) for the density matrix are established within a long time, owing to the action of relaxation processes. The initial state is therefore mixed. We shall show nevertheless that to calculate the final state of the ions, which obviously is also mixed, we can use the Schrödinger equation if, in accord with the assumptions made, the time of turning on the field is considerably shorter than the relaxation time.

Indeed, neglecting relaxation, the solution of the equation for the density matrix $\rho_V(t)$ in the interaction representation,

$$\begin{aligned}i\frac{\partial\rho_V}{\partial t}&=[\mathbf{H}\times\rho_V],\\ \rho_V&=\begin{pmatrix}\rho_{11} & \rho_{12}e^{i\delta t}\\ \rho_{21}e^{-i\delta t} & \rho_{22}\end{pmatrix},\end{aligned} \tag{7}$$

is expressed in terms of the $\mathbf{S}$ operator:

$$\begin{aligned}\rho_V(t)&=\mathbf{S}(t)\rho_V(-\infty)\mathbf{S}^+(t),\\ i\dot{\mathbf{S}}&=\mathbf{H}\mathbf{S},\quad \mathbf{S}(-\infty)=\mathbf{I}.\end{aligned} \tag{8}$$

Comparing Eqs. (1), (2), and (7) we easily obtain an explicit expression for the Hamiltonian $\mathbf{H}$:

$$\mathbf{H}=\frac{I}{2}\begin{pmatrix} -\alpha-\beta\xi_2 & (\lambda\xi_3+i\varkappa\xi_1)e^{-i\delta t} \\ (\lambda\xi_3-i\varkappa\xi_1)e^{i\delta t} & \alpha+\beta\xi_2 \end{pmatrix}.$$

If we write now the S matrix in the form

$$\mathbf{S}(t)=\begin{pmatrix} a_{11}(t) & a_{12}(t) \\ a_{21}(t) & a_{22}(t) \end{pmatrix}, \tag{9}$$

the equations for the four coefficients a_{ij} obtained from (8) break up into two independent pairs, each of which has the same form of the Schrödinger equation for a two-level system:

$$\begin{aligned} i\dot{a}_{1j}&=-{}^1/_2(\alpha+\beta\xi_2)Ia_{1j}+{}^1/_2(\lambda\xi_3+i\varkappa\xi_1)Ie^{-i\delta t}a_{2j}, \\ i\dot{a}_{2j}&={}^1/_2(\lambda\xi_3-i\varkappa\xi_1)Ie^{i\delta t}a_{1j}+{}^1/_2(\alpha+\beta\xi_2)Ia_{2j}. \end{aligned} \tag{10}$$

Equations (10) must be solved with the initial conditions

$$a_{ij}(-\infty)=\delta_{ij}.$$

We now specify concretely the form of the envelope $I(t)$. We choose it, following Ref. 6, by implicitly specifying a function $I(t)$ that permits an analytic solution of Eqs. (10):

$$t=\tau[(1-I/I_0)^{-1}-\ln(I_0/I-1)]. \tag{11}$$

The function $I(t)$ is an increasing one, with a characteristic turning-on time τ and with an asymptotic value of the amplitude I_0 (see Fig. 2 of Ref. 6). $I(t)\to I_0$ as $t\to\infty$. In this case the S matrix is expressed in terms of confluent hypergeometric functions:

$$a_{11}(t)=e^{i\varepsilon_1 x}\Phi\left[i\frac{\eta}{2}\left(1+\frac{\Delta}{\Omega}\right), i\eta; i\Omega x\right],$$

$$a_{12}(t)=\frac{i}{1-i\eta}\frac{\gamma\tau I_0}{2}\left(\frac{x}{\tau}\right)^{1-i\eta}e^{i(\varepsilon_2-\delta)x}\Phi\left[1-i\frac{\eta}{2}\left(1-\frac{\Delta}{\Omega}\right), 2-i\eta; i\Omega x\right],$$

$$a_{21}(t)=\frac{i}{1+i\eta}\frac{\gamma\tau I_0}{2}\left(\frac{x}{\tau}\right)^{1+i\eta}e^{i(\varepsilon_1+\delta)x}\Phi\left[1+i\frac{\eta}{2}\left(1+\frac{\Delta}{\Omega}\right), 2+i\eta; i\Omega x\right], \tag{12}$$

$$a_{22}(t)=e^{i\varepsilon_2 x}\Phi\left[-i\frac{\eta}{2}\left(1-\frac{\Delta}{\Omega}\right), -i\eta; i\Omega x\right],$$

$$\eta=\delta\tau,\quad \Delta=\delta+(\alpha+\beta\xi_2)I_0,\quad \gamma=|\lambda\xi_3+i\varkappa\xi_1|,$$
$$\Omega=[\Delta^2+(\gamma I_0)^2]^{1/2},\quad \varepsilon_{1,2}=-\Omega/2\pm[(\alpha+\beta\xi_2)I_0-\delta/2].$$

The function $x(t)$ is defined by the expression

$$t=x+\tau\ln(x/\tau). \tag{13}$$

The large t (i.e., at $t\gg\tau$) we have $x(t)\approx t$. Using this fact, as well as the asymptotic form of the confluent hypergeometric function[8] and expressions (8) and (12), we can obtain the asymptotic value of the magnetization of the crystal at large t. This expression is a sum of two terms—constant and oscillating in time at a frequency approximately equal to the Rabi frequency. The amplitude of the oscillating term, which we shall not write out here, decreases rapidly with increasing characteristic switching parameter $\eta=\delta\tau$. Bearing in mind, furthermore, that the Rabi oscillations average out rapidly as a result of various relaxation processes and of the spatial inhomogeneity of the field, we obtain the magnetic-sublevel population difference w, defined by a time-constant term in the form

$$w=w(-\infty)(\Delta/\Omega)(\operatorname{ch}\pi\eta-e^{-\pi\eta\Delta/\Omega})/\operatorname{sh}\pi\eta. \tag{14}$$

Assuming hereafter that the interaction is turned on slowly,[1)] i.e., $\delta>0$ and $\eta\gg1$, we obtain from (14) for the magnetization the expression

$$\begin{aligned} M_z&=M_z^{(0)}(\Delta/\Omega)\{1-2\exp[-\pi\eta(1+\Delta/\Omega)]\}, \\ M_z^{(0)}&=N\delta(\mu_{mz}-\mu_{nz})/Q\Theta, \end{aligned} \tag{15}$$

where $M_z^{(0)}$ is the initial magnetization of the crystal. As seen from (15), the crystal magnetization is determined by the product of M_z^0 and two factors, Δ/Ω and $\{\cdots\}$. The first is the magnetization in a monochromatic field I_0 following adiabatic turning-on of the interaction. The second factor yields in the limit, as will be shown below, the Landau-Zener formula for the probability of excitation in level crossing.

We consider now the behavior of the magnetization at different parameters of the problem, using concrete examples.

3. TRIPLET GROUND STATE

If the ground state of the impurity ions is a triplet, as is the case, e.g., for the paramagnetic crystals MgO: Fe^{2+}, the crystal magnetization is determined by the difference between the populations of the magnetic sublevels $m=1$ and $m=-1$. In this case, according to the selection rules (see Ref. 2), we have $\alpha=0$ and $\Delta=\delta+\beta\xi_2 I_0$.

The effective Hamiltonian of the interaction of the radiation and of the constant magnetic field with the ions is

$$\begin{aligned} H=&-{}^1/_4IT_{\Gamma_1}-{}^1/_2\Delta S_z+{}^1/_4IT_{\Gamma_3}(3S_z^2-2)+{}^1/_4iI\varkappa\xi_1(S_+^2-S_-^2) \\ &+{}^1/_4I\lambda\xi_3(S_+^2+S_-^2). \\ T_{\Gamma_3}=&\,3^{-1/2}\langle 1|T_{\Gamma_3\theta}|1\rangle. \end{aligned}$$

Here $S_\pm=S_x\pm S_y$ and S_z are spin operators, T_{Γ_1} is a parameter that determines the shift of the level as a whole. The remaining parameters are defined in (3) and (4). The magnetic-sublevel energies determined by this Hamiltonian are

$$E_\pm={}^1/_4I(T_{\Gamma_3}-T_{\Gamma_1})\pm{}^1/_2[(\delta+\beta\xi_2 I)^2+(\gamma I)^2]^{1/2}. \tag{16}$$

It follows from (16) that linearly polarized radiation ($\xi_2=0$) only pushes apart the magnetic sublevels $m=\pm1$ and inversion of their populations is impossible. For the asymptotic crystal magnetization produced after completion of the external-field transient we obtain from (15) at $\xi_2=0$ the usual result corresponding to adiabatic slow turning-on of the field:

$$M_z=M_z^{(0)}\delta[\delta^2+(\varkappa^2\xi_1^2+\lambda^2\xi_3^2)I_0^2]^{-1/2}. \tag{17}$$

It follows from (17) that when the radiation intensity is in-

creased the crystal magnetization vanishes. This is due to equalization of the populations of the sublevels $m = \pm 1$ as a result of Raman scattering of the radiation by the impurity ions. The magnetization is subsequently restored because of relaxation processes.

If the radiation frequency ω is close to the frequency of the dipole transition of the electron in the impurity ion, the parameters β, $\varkappa$, and λ satisfy in the approximation with one intermediate level the simple relations $\varkappa^2 = \beta^2 = \lambda^2$, if the intermediate level is not the doublet $\Gamma/3$, and $\varkappa^2 = \beta^2 = \lambda^2/4$ in the opposite case.

In the first case, the crystal magnetization (17) does not depend on the orientation of the radiation polarization plane. In the second case, if the radiation intensity I_0 remains unchanged, the crystal magnetization is a minimum if the polarization plane is parallel to one of the symmetry axes of the cubic crystal ($\xi_1 = 0$, $\xi_3 = 1$).

We consider now a case when elliptically polarized radiation passes through the crystal. The behavior of the levels is determined, according to (16), by the relation between the degrees of linear and circular polarizations, and at appropriate values of the problem parameters the levels can come together and intersect. From expression (16) for the sublevel magnetizations we can estimate the radiation intensity at which the sublevel come closest together. For a magnetic field $B \sim 100$ G we have $I_0 \sim 10^8$ W/cm^3.

The behavior of the magnetization is determined by the relations between the problem parameters γ, I_0, Δ, Ω, and η. The expression for the magnetization is

$$M_z = M_z^{(0)}\,\Delta/\Omega \qquad (18)$$

at $\eta \gg 1$, $\eta(1 + \Delta/\Omega) \gg 1$. The second inequality means either the absence of level crossing or that γI_0 is not small compared with $|\Delta|$. For example, under conditions of resonance with a dipole transition in the impurity ion this condition is satisfied if the degree of linear polarization of the radiation is not too small. If, however $\eta \gg 1$, $\eta(1 + \Delta/\Omega) \lesssim 1$, as can be the case when the sublevels cross and, in addition, at small $|\gamma I_0/\Delta|$, we have

$$M_z = M_z^{(0)}\,\{2\exp[-{}^1/_8\pi\eta(\gamma I_0)^2] - 1\}. \qquad (19)$$

Equation (19) corresponds to the Landau-Zener approximation as applied to our problem. Under conditions of resonance with a dipole transition in the impurity ion, this equation takes the form

$$M_z = M_z^0\{2\exp[-\pi\eta(1-\xi_2^2)\beta^2 I_0^2/(\delta+\beta\xi_2 I_0)^2] - 1\}. \qquad (20)$$

The magnetization is determined here by the degree of circular polarization of the radiation.

It can be seen that Eqs. (18) and (19) admit of the possibility of change of the sign of the magnetization up to total inversion: $M_z = -M_z^0$. Figure 2 shows the behavior of the magnetization as a function of the field intensity I_0 in dimensionless units, for different values of the degree of circular polarization ξ_2. In accordance with the statements made above for linear polarization ($\xi_2 = 0$) we have $M_z \to 0$ with increasing I_0 (curve 1). At $\xi_2 < 0$ a possibility arises of cross-

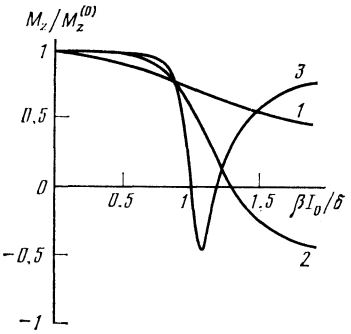

FIG. 2. Magnetization of a crystal in a ground triplet state vs the radiation intensity: 1—$\xi_2 = 0$; 2—$\xi_2 = 0.9$; 3—$\xi_2 = -0.999$. For all the curves $\pi\eta = 5$.

ing of the magnetic sublevels at $I_0 = I_c$, where I_c corresponds to the Stark shift, which is equal to δ:

$$I_c = -\delta/\beta\xi_2.$$

At $I_0 > I_c$ the magnetization can reverse sign. The parameter $\pi\eta(1 + \Delta/\Omega)$ for curve 2 is equal to 4, which corresponds to Eq. (18) that describes adiabatic inversion of the levels. If the parameter ξ_2 approaches -1, the behavior of $M_z(I_0)$ changes (see curve 3). At such values of ξ_2 we have $\pi\eta(1 + \Delta/\Omega) \lesssim 1$ and the magnetization is described by the Landau-Zener formula (19). It can be seen from Fig. 2 that the magnetization-inversion region is the section $I_c < I_0 < I'_c$. At $I_0 < I_c$ and $I > I'_c$ there is no inversion. This $M_z(I_0)$ dependence is understandable. At $I_0 < I_c$ there is still no crossing, and at $I_0 > I'_c$ a nonadiabatic jump through resonance takes place—the level-crossing time is too short for a transition to take place. The value of I'_c is easy to estimate. For relation (20) it is equal to

$$I_c' = (\delta/\beta)\{\xi_2 + [\pi\eta(1-\xi_2^2)/\ln 2]^{1/2}\}^{-1}.$$

The dependence of the magnetization on the degree of circular polarization at various field intensities is shown in Fig. 3. It can be seen that for each I_0 the magnetization is reversed in a definite region of values of ξ_2, i.e., at $\xi_2^{(1)} < \xi_2 < \xi_2^{(0)}$. The value $\xi_2^{(0)}$ corresponds to a polarization at which I_0 is equal to I_c (and then $\Delta = 0$), while $\xi_2 < \xi_2^{(1)}$ corre-

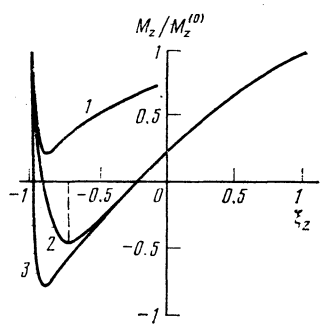

FIG. 3. Crystal magnetization in a ground triplet state vs the degree of circular polarization of the radiation at a fixed intensity: 1—$\pi\eta = 5$, $\beta I_0/\delta = 1$; 2—$\pi\eta = 5$, $\beta I_0/\delta = 6$; 3—$\pi\eta = 20$, $\beta I_0/\delta = 6$. The dashed line marks the value of $\xi_2^{(m)}$ for curve 2.

sponds to a nonadiabatic jump through the resonance. There is thus a circular polarization $\xi_2^{(m)}$ corresponding to the maximum inversion. Estimates for the first case considered above (the intermediate level, Γ_3) yield

$$\xi_2^{(m)} \approx 1 - 2\frac{\ln 2\pi\eta}{\pi\eta}\frac{(\delta-\beta I_0)^2}{\beta^2 I_0^2}.$$

In the more general case of arbitrary relations between the parameters β, $\varkappa$, and λ the main features of the behavior of the magnetization remain the same, but the dependence on the radiation polarization becomes more complicated.

4. QUARTET GROUND STATE

Crystals in which the ground state of the impurity ions is are quartet are, e.g., Cs_2ZrCl_6: Pa^{4+}, MgO: Er^{3+}, CaF_2: Pr^{2+}. In this case the crystal magnetization is determined by the sum of magnetizations of two pairs of magnetic sublevels, (3/2, − 1/2) and (1/2, − 3/2), which are mixed by the optical radiation. In this case

$$M_z = \frac{1}{2}M_z^{(0)}\left\{\frac{\Delta_{3/2}}{\Omega_{3/2}}\left[1-2\exp\left\{-\pi\eta\left(1+\frac{\Delta_{3/2}}{\Omega_{3/2}}\right)\right\}\right]\right.$$
$$\left.+\frac{\Delta_{1/2}}{\Omega_{1/2}}\left[1-2\exp\left\{-\pi\eta\left(1+\frac{\Delta_{1/2}}{\Omega_{1/2}}\right)\right\}\right]\right\}.$$

Here

$$\Delta_{3/2,\,1/2} = \delta + (\xi_2\beta_{3/2,\,1/2} \pm \alpha_{3/2})I_0,$$
$$\Omega_{3/2,1/2}^2 = \Delta_{3/2,1/2}^2 + (\xi_1^2\varkappa_{3/2,1/2}^2 + \xi_3^2\lambda_{3/2,1/2}^2)I_0^2$$

are the resultant detuning from resonance and the Rabi frequency for the sublevel pairs (3/2, − 1/2) and (1/2, − 3/2) respectively. The character of the dependence of each term on the radiation parameters was investigated in the preceding section. A feature of this case is that the level crossing in these pairs sets in, generally speaking, not simultaneously, and the parameter $\pi\eta(1+\Delta/\Omega)$, which determines the behavior of the value of each term, can have different values corresponding to different regimes of inversion (or absence of inversion) for each sublevel pair.

Let us consider in greater detail the concrete situation of an intermediate resonant doublet. The matrix elements β, $\varkappa$, and λ satisfy here the relations[2] $\varkappa^2 = \lambda^2 = \frac{4}{3}\beta^2$. If both parameters $\pm\alpha + \beta\xi_2 \geqslant 0$, there is no crossing and the magnetization tends to zero or to a certain equilibrium value as $I_0 \to \infty$. For noticeable magnetization inversion it is necessary that the crossing take place in both doublets (3/2, − 1/2) and (1/2, − 3/2), i.e., at $\pm\alpha + \beta\xi_2 > 0$. Therefore in plane-polarized light ($\xi_2 = 0$) inversion takes place in only one doublet and the magnetization reversal effect is noticeably lowered by the second pair of sublevels. The situation is similar if $\alpha > \beta\xi_2$. The $M_z(I_0)$ dependence for this case is shown in Fig. 4a. It can be seen that inversion takes place for the level pair (1/2, − 3/2), and the total magnetization does not reverse sign.

In addition to presence of crossing in both doublets, magnetization reversal calls for the crossing of the inversion region to take place on the I_0 axis and that the condition

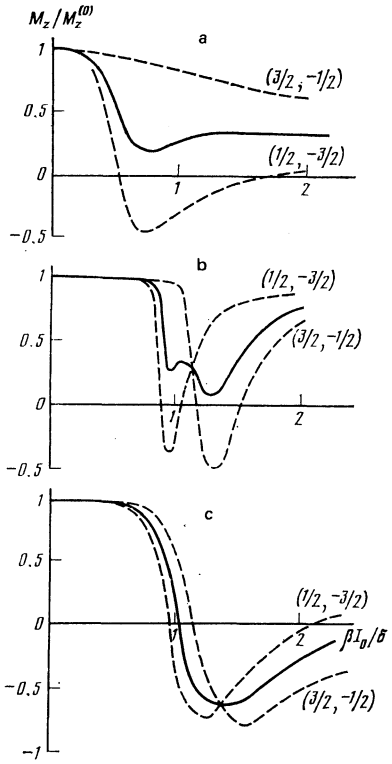

FIG. 4. Dependence of the magnetization on the radiation intensity for crystals with impurity ions whose ground state is a quartet (solid line). The dashed lines mark the dependences of double the values of the magnetic moment of the component doublets: a—$\xi_2 = -0.9$, $\alpha/\beta = 1$, $\pi\eta = 20$; b—$\xi_2 = -0.999$, $\alpha/\beta = 0.15$, $\pi\eta = 20$; c—$\xi_2 = -0.99$, $\alpha/\beta = 0.1$, $\pi\eta = 20$.

$\pi\eta(1+\Delta/\Omega) \lesssim 1$ be satisfied at least for one of the doublets. This means that the inequality $\alpha \ll \beta\xi_2$ must be satisfied for one of the doublets. It can be seen from Fig. 4(b) that despite the presence of doublets in both doublets, the total magnetization does not reverse sign. The curve on Fig. 4(c) corresponds to satisfaction of the two aforementioned conditions. We have here adiabatic inversion for both problem in approximately the same region of values of I_0, and consequently the total magnetization reverses sign.

We can analyze similarly the dependence of the total magnetization on the degree of circular polarization; a typical plot of $M_z(\xi_2)$ is similar to that shown in Fig. 3. Noticeable magnetization inversion takes place only if the inversion regions on the axis are approximately the same for both doublets. In analogy with the case of triplet levels, there is a degree of circular polarization $\xi_2^{(m)}$ at which the magnetization is a maximum.

5. INHOMOGENEOUS BROADENING

The foregoing analysis corresponds to ideal crystals in which all the impurity ions are under identical conditions. However, by virtue of the inhomogeneity of the impurity-ion distribution in the crystal and of the crystal-lattice distortions, the local fields are different for different ions. In first-order approximation this phenomenon is usually described by introducing inhomogeneous broadening. We assume

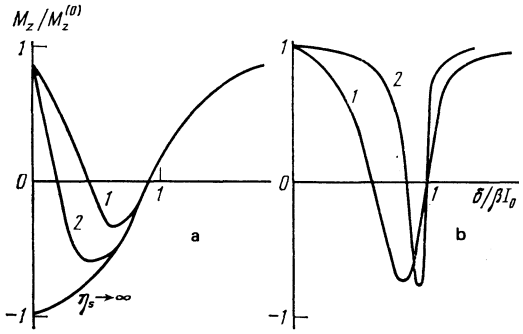

FIG. 5. Magnetization of an ideal crystal with triplet ground level of the ions vs the splitting δ: a)—adiabatic regime, 1—$\xi_2 = -0.9$, $\pi\eta_s = 5$, 2—$\xi_2 = -0.9$, $\pi\eta_s = 20$; b)—Landau-Zener regime of inversion 1—$\pi\eta_s = 20$, $\xi_2 = -0.99$; 2—$\pi\eta_s = 20$, $\xi_2 = -0.999$.

hereafter that the inhomogeneous broadening is due to inhomogeneity of the magnetic field **B** in the crystal, and since the constant magnetic field enters via the detuning δ, inhomogeneity of **B** corresponds to a distribution of δ in accord with a certain law near a mean value δ_0. For the sake of argument we put $\delta_0 > 0$. Usually the inhomogeneous broadening is approximated by a Gaussian distribution. We thus obtain for the magnetization of the crystal

$$\langle M_z\rangle = \int_{-\infty}^{\infty} g(\delta-\delta_0) M_z(\delta)\, d\delta, \tag{21}$$

$$g(\delta-\delta_0) = \pi^{-1/2} T_2 \exp\left[-(\delta-\delta_0)^2 T_2^2\right], \tag{22}$$

where T_2^{-1} is the magnitude of the inhomogeneous broadening.

We note that if T_2^{-1} is comparable in magnitude with the sublevel splitting δ_0 in the magnetic field, it is incorrect to use Eq. (21), since it takes into account only the local field, but not its spatial orientation, which becomes an important factor in the case of a large inhomogeneous width. We shall assume hereafter that $\delta_0 T_2 \gg 1$. It can be seen from (21) that the resultant magnetization of the crystal is determined by the integral of the overlap of the areas under the distribution line and the dispersion curve that gives the dependence of a magnetic ideal crystal on the sublevel splitting, $M_z(\delta)$. We consider this dependence in greater detail, confining ourselves to the triplet case.

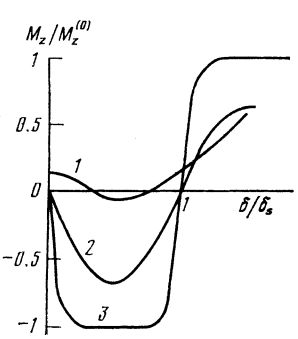

FIG. 6. Crystal magnetization with allowance for inhomogeneous broadening: 1—$\delta_s T_2 = 1$; 2—$\delta_s T_2 = 2$; 3—$\delta_s T_2 = 10$.

The dependence of M_z on δ, just as on I_0, is determined by the relations between the parameters of the problems. Since we are interested in the case of level crossing, we assume that $\xi_2 < 0$. Of importance for the dependence of M_z on δ is the parameter $\eta_s = \delta_s \tau$ (where δ_s is the resultant Stark shift of the sublevels), which plays the role of the adiabaticity parameter. At $\eta_s \gg 1$ it is possible to separate in the dependence of M_z on ξ_2 two characteristic regimes, adiabatic at $\eta_s(1+\Delta/\Omega) \gg 1$ with a magnetization maximum at $\delta \sim 0$, and a Landau-Zener regime if $\eta_s(1+\Delta/\Omega) \lesssim 1$, with a maximum of the magnetization at $\delta = -\xi_2\delta_s$. The dispersion curves corresponding to these regimes are shown in Fig. 5. Understandably, if η_s is at the boundary of the inversion region, the inhomogeneous broadening can substantially decrease or cover up completely the magnetization inversion.

To calculate $\langle M_z$ it is necessary to substitute in (21) the formula (14) for $M_z(\delta)$. The resultant integral can be calculated only numerically. We shall consider the role of the inhomogeneous broadening in the limiting case of the adiabatic inversion regime, when $\eta_s \gg 1$ and $\xi_2 \approx -1$, so that

$$M_z(\delta) = M_z^{(0)} \frac{\Delta}{\Omega} \approx \begin{cases} M_z^{(0)}, & \delta<0, \quad \delta>-\xi_2\delta_s, \\ -M_z^{(0)}, & 0<\delta<-\xi_2\delta_s. \end{cases}$$

The magnetization can thus be represented by a step function. The integral (21) can here be calculated exactly:

$$\langle M_z\rangle = M_z^{(0)}\{1-\Phi(\delta_0 T_2) - \Phi[(\delta_s-\delta_0)T_2]\}.$$

Here $\Phi(x)$ is the probability integral.[8]

Figure 6 shows the dependence of the magnetization $\langle M_z\rangle/M_z^{(0)}$ on δ_0/δ_s. It can be seen that if $\delta_s T_2 \lesssim 1$, i.e., the Stark shift is within the limits of the inhomogeneous width, the effect of magnetization inversion becomes strongly smeared out, since there is no inversion on the wings of the inhomogeneous line. If, however, $\delta_s T_2 > 1$, the inhomogeneous broadening smooths out the step. The width of the smearing region is $\sim T_2^{-1}$. The maximum magnetization reversal of the crystal is in the region $\delta_0 \approx \delta_s/2$ and amounts to

$$\langle M_z\rangle \approx M_z^{(0)}[1-2\Phi(\delta_s T_2/2)].$$

Since $\Phi(2) \approx 1$, we can state that total reversal of the crystal magnetization can be obtained already at $\delta_s T_2 \approx 4$ and when the conditions for the inversion of the sublevel populations is satisfied. An estimate of the radiation intensity needed to reverse the magnetization yields $I_0 \sim 10^8$ W/cm^2 at a characteristic inhomogeneous broadening $T_2^{-1} \lesssim 0.1$ cm^{-1}.

[1)]In magnetic fields $B \sim 10^4$ G this condition is satisfied at $\tau > 10^{-11}$ sec.

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