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Nonlinear paramagnetic Faraday effect

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A microscopic theory of the nonlinear Faraday effect is developed for cubic crystals containing paramagnetic impurity ions. The autorotation and deformation of the polarization ellipse are calculated for strong electromagnetic radiation propagated in such crystals. It is shown that a change of polarization of the radiation occurs even for ions whose ground state is a "nonmagnetic" doublet, for which the ordinary paramagnetic Faraday effect is absent. Allowance is made for the effect of the change of polarization of the wave on the magnetization of the crystal, and this permits refinement of Pershan's theory of the inverse Faraday effect. Nonstationary phenomena are considered, and it is shown that a relaxationless change of the crystal magnetization by a short light pulse is possible by virtue of the combinational light scattering that occurs between magnetic sublevels of the ions, split by a constant magnetic field.

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1. INTRODUCTION

In a magnetic field, degenerate states of paramagnetic ions are split and, at not very high temperatures, are occupied with unequal probabilities. Consequently a crystal with paramagnetic impurities possesses circular birefringence, which leads to a Faraday rotation of the plane of polarization of radiation propagated along the magnetic field. The amount of this rotation is proportional to the magnetic field and inversely proportional to the temperature. But if the radiation is sufficiently strong, it also perturbs the spectrum of the ions, and this leads to a dependence of the Faraday rotation on the intensity of the radiation.

Besides the effect on the index of refraction, elliptically polarized radiation leads also to magnetization of the medium. This phenomenon is known as the inverse Faraday effect (IFE)[1] and is similar to the rectification effect known for ferrites.[2] The terminological difference is due to the fact that the IFE is observed in the optical range and is caused by interaction with the electric vector of the wave, whereas the rectification effect observed in the radiofrequency range is caused by magnetic interaction.

The nonlinear corrections to the Faraday rotation and the IFE are closely related to each other. In the present paper, a microscopic treatment of both these phenomena is given for cubic paramagnetic crystals. In Ref. 1, the IFE was treated phenomenologically and without allowance for the change of the polarization characteristics of the wave during is propagation. Also, no analysis was made of the nonstationary phenomena that occur for pulsed electromagnetic radiation, although the existing experimental methods of observation of the IFE are based precisely on the pulsed character of the phenomenon.[3,4] Furthermore, the possibility of the presence of a constant external magnetic field leads to new features of the IFE, connected with a change of the populations of the magnetic sublevels as a result of the process of combinational (Raman) scattering (CS).[5]

In section 2, the effective Hamiltonian is given for interaction of an ion with external fields. In section 3, the kinetic equation for an ion is obtained in the relaxation approximation. In section 4, the permittivity tensor of a paramagnetic crystal is calculated with allowance for nonlinear effects, and a closed system of equations is obtained for the field and the material, with allowance for the change of polarization of the radiation during propagation in the crystal. These equations are solved in the subsequent sections: in section 5 for ions whose ground state is a Kramers doublet (in this case the equations can be solved exactly for an arbitrary variation of the radiation intensity with time), and in section 6 for ions in nonmagnetic doublet, triplet, or quadruplet states (for the case of a slow variation of the intensity with time, in comparison with the paramagnetic relaxation times). The results obtained make it possible to calculate the nonlinear corrections to the paramagnetic Faraday effect and are used in section 7 to calculate the IFE. In section 8, the IFE is calculated for a short light pulse, when relaxation processes are unimportant and CS is the determining mechanism of magnetization of the crystal.

The Appendix presents some properties of the irreducible tensor operators of the cubic symmetry group; these are necessary for carrying out the calculation. Throughout the paper, the Planck constant $\hbar$ and the Boltzmann constant are set equal to unity.

2. THE EFFECTIVE HAMILTONIAN

Let a plane quasimonochromatic wave, for which the real value of the electric field is

$$\mathbf{E}(z,t)=\operatorname{Re}\{\mathbf{E}_0(z,t)\exp[i(kz-\omega t)]\},$$

be propagated along a principal crystallographic axis of a cubic crystal, which we take as the Z axis. A constant magnetic field $\mathbf{B}$ is also directed along this axis. If the frequency ω of the radiation is sufficiently high, the perturbation of the spectrum of the ions reduces to a dynamic quadratic Stark effect. The perturbation of the spectrum is strongest when ω is close to a characteristic absorption frequency of the ion.1) The perturbation of its crystalline environment may then be neglected.

In the dipole approximation, the effective Hamiltonian describing the interaction of the ion with the fields $\mathbf{B}$ and $\mathbf{E}$ has the form[6]

$$H=-\boldsymbol{\mu}\mathbf{B}-\frac{1}{4}I(z,t)\left\{T_{\Gamma_1}-\frac{1}{\sqrt{3}}T_{\Gamma_3\theta}+\xi_3T_{\Gamma_3\varepsilon}+\xi_2T_{\Gamma_40}+\xi_1T_{\Gamma_50}\right\}. \tag{1}$$

Here $\boldsymbol{\mu}$ is the magnetic moment operator, and $I=\mathbf{E}_0\cdot\mathbf{E}_0^*$ is the intensity of the acting alternating field. The Stokes parameters are determined by the formulas

$$\xi_1-i\xi_2=2E_{0x}E_{0y}^*/I,\quad \xi_3=(|E_{0x}|^2-|E_{0y}|^2)/I.$$

We recall that ξ_2 determines the degree of circular polarization and $(\xi_1^2+\xi_3^2)^{1/2}$ the degree of linear polarization. The parameter ξ_3 characterizes the linear polarization along the crystal axes, ξ_1 along axes rotated with respect to the crystal axes through an angle $\pi/4$. The radiation is assumed to be completely polarized, so that $\xi_1^2+\xi_2^2+\xi_3^2=1$.

The operators T in (1) have the following form:

$$T_{\Gamma_1}={}^1/_3(D_{xx}^{(+)}+D_{yy}^{(+)}+D_{zz}^{(+)}),\quad T_{\Gamma_40}=iD_{xy}^{(-)},\quad T_{\Gamma_50}=D_{xy}^{(+)},$$

$$T_{\Gamma_3\theta}=\frac{1}{2\sqrt{3}}(2D_{zz}^{(+)}-D_{xx}^{(+)}-D_{yy}^{(+)}),\quad T_{\Gamma_3\varepsilon}=\frac{1}{2}(D_{xx}^{(+)}-D_{yy}^{(+)}),$$

$$D_{ij}^{(\pm)}={}^1/_2\{d_i(G^{(-)}\pm G^{(+)})d_j\pm d_j(G^{(-)}\pm G^{(+)})d_i\},$$

$\mathbf{d}$ is the electric dipole moment operator, and the operators $G^{(\pm)}$ are determined by a sum over all excited states $|\lambda\rangle$ of the ion:

$$G^{(\pm)}=\sum_\lambda\frac{|\lambda\rangle\langle\lambda|}{\omega_{\lambda0}\pm\omega},\quad \omega_{\lambda0}=E_\lambda-E_0, \tag{2}$$

where $E_{\lambda,0}$ are the unperturbed energies.

The indices of the operators $T_{\Gamma M}$ denote the irreducible representation of the cubic group according to which these operators transform. We note that the part of the Hamiltonian (1) proportional to the intensity of the radiation leads to splitting of a level only in the presence of orbital degeneracy.

The state of an ion is conveniently characterized by an effective spin S and its projection m.[7] For the Kramers doublets $\Gamma_{6,7}$, $S=\frac{1}{2}$ and $m=\pm\frac{1}{2}$. For the triplets $\Gamma_{4,5}$, $S=1$ and $m=0,\pm1$; for the quadruplets Γ_8, $S=\frac{3}{2}$ and $m=\pm\frac{3}{2},\pm\frac{1}{2}$. The nonmagnetic doublet Γ_3 we shall characterize by effective spin $S=2$ and its sublevels by ε and θ.

The values of the magnetic elements of the operators $T_{\Gamma M}$ that are needed for the present paper are given for states $|Sm\rangle$ in the Appendix.

3. THE KINETIC EQUATION

By supposing that only sublevels of the ground state of the ion are occupied and by using methods developed in the theory of magnetic relaxation,[8] one can obtain the equation for the density matrix

$$\frac{\partial\rho}{\partial t}=i[\rho,H]+R(\rho^{(0)}-\rho), \tag{3}$$

where R is a relaxation operator, and where $\rho^{(0)}$ is the equilibrium density matrix, determined by the expression

$$\rho^{(0)}(t)=\exp(-H/\Theta)/\operatorname{Sp}[\exp(-H/\Theta)],$$

Θ is the temperature of the crystal in energy units.

Supposing that $H/\Theta\ll1$, one can rewrite the expression for $\rho^{(0)}$ in the form

$$\rho^{(0)}(t)=\frac{1}{Q}\left[1-\frac{1}{\Theta}\left(H-\frac{1}{Q}\operatorname{Sp}H\right)\right], \tag{4}$$

where Q is the multiplicity of the ground state degeneracy. The matrix $\rho^{(0)}$ may vary with time when the radiation intensity is not constant. If τ is a characteristic time of variation of the function $I(t)$, then formulas (3) and (4) hold when the inequality

$$\tau\gg\tau_c, \tag{5}$$

is satisfied, where τ_c is the correlation time of the lattice. For typical crystals, $\tau_c\sim10^{-12}-10^{-13}$ sec.[9] The relation (5) means that the lattice, which in this case plays the role of a thermostat, changes its state rapidly, forming an equilibrium state of the ion for each new value of the radiation intensity.

When, along with (5), the relation $H\tau_c\ll1$ is also satisfied, the relaxation times are independent of the field intensities. Introducing, in accordance with general custom, longitudinal and transverse relaxation times T_1 and T_2, we write the operator R in the form

$$R_{ij}(\rho^{(0)}-\rho)=\begin{cases}(\rho_{ii}^{(0)}-\rho_{ii})/T_1, & i=j\\ (\rho_{ij}^{(0)}-\rho_{ij})/T_2, & i\neq j\end{cases}.$$

In the Hamiltonian (1), only the operators $T_{\Gamma_3\varepsilon}$ and T_{Γ_50} possess nondiagonal matrix elements, connecting the states m and $m\pm2$ of the triplets and quadruplets and the states θ and ε of the doublet Γ_3. Consequently, in the system (3) no more than two sublevels at a time are coupled, to which correspond three equations. In general, equations (3) can be rewritten in the following manner:

$$\begin{aligned}\partial\rho_{mm}/\partial t&=i(\rho_{mn}H_{nm}-\rho_{mn}^{*}H_{nm}^{*})+(\rho_{mm}^{(0)}-\rho_{mm})/T_1,\\ \partial\rho_{nn}/\partial t&=-i(\rho_{mn}H_{nm}-\rho_{mn}^{*}H_{nm}^{*})+(\rho_{nn}^{(0)}-\rho_{nn})/T_1,\\ \partial\rho_{mn}/\partial t&=i[(H_{nn}-H_{mm})\rho_{mn}-H_{mn}(\rho_{mm}-\rho_{nn})]+(\rho_{mn}^{(0)}-\rho_{mn})/T_2.\end{aligned}\tag{6}$$

Here m and n take the following values: $m=\theta$, $n=\varepsilon$ for the doublets Γ_3; $m=-n=1$ for the triplets; $m=\frac{3}{2}$ and $n=-\frac{1}{2}$ or $m=\frac{1}{2}$ and $n=-\frac{3}{2}$ for the quadruplets. For the Kramers doublets, $m=-n=\frac{1}{2}$, and the diagonal and nondiagonal elements of the density matrix satisfy independent equations, since the right side of the last equation (6) in this case disappears.

Transforming in (6) to real quantities and parametrizing the Hamiltonian H, we rewrite these equations as follows:

$$\begin{aligned}\partial u/\partial t&=\varkappa\xi_1 Iw-\Delta v+(\lambda\xi_3 I-u)/T_2,\\ \partial v/\partial t&=-\lambda\xi_3 Iw+\Delta u+(\varkappa\xi_1 I-v)/T_2,\\ \partial w/\partial t&=(\lambda\xi_3 v-\varkappa\xi_1 u)I+(\Delta-w)/T_1,\end{aligned}\tag{7}$$

where

$$u=2Q\Theta\,\mathrm{Re}\,\rho_{mn},\quad v=2Q\Theta\,\mathrm{Im}\,\rho_{mn},\quad w=Q\Theta(\rho_{mm}-\rho_{nn}),$$

$$\varkappa=\frac{1}{2i}\langle m|T_{\Gamma_5 0}|n\rangle,\quad \lambda=\frac{1}{2}\langle m|T_{\Gamma_3\varepsilon}|n\rangle,$$

$$\Delta=\langle m|\mu_z B+\frac{1}{4}I\left(\xi_2 T_{\Gamma_4 0}-\frac{1}{\sqrt{3}}T_{\Gamma_3\theta}\right)|m\rangle$$

$$-\langle n|\mu_z B+\frac{1}{4}I\left(\xi_2 T_{\Gamma_4 0}-\frac{1}{\sqrt{3}}T_{\Gamma_3\theta}\right)|n\rangle.$$

The three equations of the system (7) determine the difference of populations of the magnetic sublevels. The sum of the populations of the sublevels of the doublets remains constant. The magnetic sublevel $|0\rangle$ of a triplet does not display magnetooptical activity; therefore for the triplets, it is sufficient to know the difference of populations of the sublevels $|\pm 1\rangle$. But for the quadruplets it is necessary to know the relative population of each pair of sublevels,

$$\sigma=4\Theta(\rho_{3/2,\,3/2}+\rho_{-1/2,\,-1/2}-\rho_{1/2,\,1/2}-\rho_{-3/2,\,-3/2}).$$

The equation for σ is easily obtained from the first two equations (6):

$$\begin{gathered}\partial\sigma/\partial t=(\sigma_0-\sigma)/T_1,\quad \sigma_0=2[(\langle{}^3/_2|\mu_z|{}^3/_2\rangle\\ +\langle-{}^1/_2|\mu_z|-{}^1/_2\rangle)B+{}^1/_4\xi_2 I(\langle{}^3/_2|T_{\Gamma_4 0}|{}^3/_2\rangle+\langle-{}^1/_2|T_{\Gamma_4 0}|-{}^1/_2\rangle)].\end{gathered}\tag{8}$$

Furthermore, for the quadruplets the values of u, v, w, and Δ in equations (7) depend on the index m, which, as was stated above, takes two values: $\frac{1}{2}$ and $\frac{3}{2}$. This index was omitted, for brevity, in (7); but it must be restored in the cases needed.

4. THE PERMITTIVITY TENSOR

Paramagnetic impurity ions make a contribution to the polarization of the crystal in accordance with the relation

$$P_i=\mathrm{Sp}(\rho d_i)=\sum_j \mathrm{Re}(\alpha_{ij}E_j),$$

where α_{ij} is the polarizability tensor. This expression may be regarded as the definition of α_{ij}. The components of the tensor α_{ij} can be expressed in terms of the operators $T_{\Gamma M}$ that occur in the effective Hamiltonian:

$$\begin{gathered}\alpha_{xx(yy)}=\mathrm{Sp}\left[\rho\left(T_{\Gamma_1}-\frac{1}{\sqrt{3}}T_{\Gamma_3\theta}\pm T_{\Gamma_3\varepsilon}\right)\right],\\ \alpha_{xy}=\alpha_{yx}^{*}=\mathrm{Sp}\,[\rho(T_{\Gamma_5 0}-iT_{\Gamma_4 0})],\quad \alpha_{xz}=\alpha_{yz}=0.\end{gathered}\tag{9}$$

The anisotropy of the permittivity is determined by the formulas

$$\begin{gathered}\varepsilon_{xx}-\varepsilon_{yy}=(16\pi N/Q\Theta)\lambda u,\\ \varepsilon_{xy}=\varepsilon_{yx}^{*}=(8\pi N/Q\Theta)[\varkappa v-i(\beta^{(+)}w+\beta^{(-)}\sigma)],\\ \beta^{(\pm)}={}^1/_4(\langle m|T_{\Gamma_4 0}|m\rangle\mp\langle n|T_{\Gamma_4 0}|n\rangle),\end{gathered}\tag{10}$$

where N is the concentration of the ions. In the case of doublets and triplets, the values of u, v, and w are determined in (7). For such levels, $\beta^{(-)}=0$, and the parameter σ drops out of equation (10). But in the case of quadruplets, the quantities that occur in (10) are

$$u=u_{3/2}+u_{1/2},\quad v=v_{3/2}+v_{1/2},\quad w=w_{3/2}+w_{1/2},$$

where the lower index determines the value of the quantum number m in equations (7), in accordance with the remark made at the end of the preceding section.

If the density matrix ρ corresponds to equally probable occupancy of the ion sublevels that are degenerate in the absence of a field, the quantities (10) disappear. In fact, in this case $w=\sigma=0$, and the parameters u and v may be regarded as random quantities, equal on the average to zero. In the presence of fields, an optically isotropic crystal acquires the properties of a biaxial gyrotropic crystal. In this connection it must be noted that under the action of an axially symmetric perturbation, a cubic crystal becomes uniaxial.[10] The biaxiality that occurs in the present case ($\varepsilon_{xx}\neq\varepsilon_{yy}$) is due to the absence of axial symmetry for the field of a plane electromagnetic wave with arbitrary polarization.

For doublet states of the ions, the anisotropy of the crystal decreases. For the doublet Γ_3, $\varkappa=\beta^{(\pm)}=0$, and the crystal is biaxial and nongyrotropic ($\varepsilon_{xy}=0$). But if the ground state of the ion is a Kramers doublet, then $\lambda=\varkappa=0$, and the crystal becomes uniaxial and gyrotropic.

The functions u, v, and w, and consequently also the quantities (10), are determined by the intensity and the Stokes parameters of the radiation at all instants of time preceding that considered. Therefore propagation of waves in a crystal with allowance for nonlinear effects leads in the general case to complicated equations. Here we shall restrict ourselves to the case in which the nonlinear effects may be considered small.

Setting the permeability equal to unity, we write the abbreviated Maxwell equations for slowly varying amplitudes:

$$k\frac{\partial E_{0i}}{\partial z}+\frac{\varepsilon_0\omega}{c^2}\frac{\partial E_{0i}}{\partial t}=\frac{i\omega^2}{2c^2}\sum_j\varepsilon_{ij}E_{0j},\quad \varepsilon_0=(kc/\omega)^2.\tag{11}$$

By introducing the variables $\zeta=t+kz/\omega$ and $\eta=t-kz/\omega$, one can derive from (11) the following equations for the Stokes parameters:

$$\begin{aligned}\frac{\partial\xi_1}{\partial\zeta}&=\frac{\omega}{2\varepsilon_0}\left[\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})\xi_2+\xi_3\,\mathrm{Im}\,\varepsilon_{xy}\right]=\frac{4\pi\omega N}{Q\Theta\varepsilon_0}[\lambda u\xi_2-(\beta^{(+)}w+\beta^{(-)}\sigma)\xi_3],\\ \frac{\partial\xi_2}{\partial\zeta}&=\frac{\omega}{2\varepsilon_0}\left[\xi_3\,\mathrm{Re}\,\varepsilon_{xy}+\frac{1}{2}(\varepsilon_{yy}-\varepsilon_{xx})\xi_1\right]=\frac{4\pi\omega N}{Q\Theta\varepsilon_0}[\varkappa v\xi_3-\lambda u\xi_1],\\ \frac{\partial\xi_3}{\partial\zeta}&=\frac{\omega}{2\varepsilon_0}[\xi_1\,\mathrm{Im}\,\varepsilon_{yx}-\xi_2\,\mathrm{Re}\,\varepsilon_{xy}]=\frac{4\pi\omega N}{Q\Theta\varepsilon_0}[(\beta^{(+)}w+\beta^{(-)}\sigma)\xi_1-\varkappa v\xi_2].\end{aligned}\tag{12}$$

Equations (12) must be solved simultaneously with equations (7). We turn now to an analysis of the solutions.

5. KRAMERS DOUBLET

For Kramers doublets, the effective Hamiltonian (1) has no nondiagonal matrix elements. Therefore the nondiagonal elements of the density matrix $\rho_{1/2,-1/2}$ do not change after application of fields. Then the phases of $\rho_{1/2,-1/2}$ may be considered random quantites, and only the diagonal elements of the density matrix make a contribution to the observed effects.

The equation for the population difference of the sublevels of a Kramers doublet is obtained from the third equation of the system (7):

$$\partial w/\partial t=(\Delta-w)/T_1, \qquad (13)$$

where

$$w=2\Theta(\rho_{1/2,\,1/2}-\rho_{-1/2,\,-1/2}), \quad \Delta=2\langle{}^1/_2|\mu_z B+{}^1/_4 I\xi_2 T_{\Gamma_4 0}|{}^1/_2\rangle.$$

Here it has been taken into account that the matrix elements of the operator $T_{\Gamma_3\theta}$ in this case vanish. Since the matrix elements of the operators T_{Γ_3} and $T_{\Gamma_5\theta}$ also vanish, we get from equation (12)

$$\partial\xi_1/\partial\zeta=-aw\xi_3, \quad \partial\xi_2/\partial\zeta=0, \quad \partial\xi_3/\partial\zeta=aw\xi_1,$$
$$a=(\pi\omega N/\Theta\varepsilon_0)\langle{}^1/_2|T_{\Gamma_4 0}|{}^1/_2\rangle. \qquad (14)$$

It follows from equations (14) that the polarization ellipse of the wave does not change during propagation of the wave in the crystal. This becomes intelligible if we notice that the anisotropy of the crystal in the case considered is caused solely by the element $\mathrm{Im}\,\varepsilon_{xy}$; this, as is well known,[10] leads only to rotation of the polarization ellipse.

It follows from the second equation (14) that the Stokes parameter ξ_2 may be an arbitrary function of the phase variable $\eta=t-kz/\omega$. If the polarization of the radiation incident on the forward plane of the crystal is independent of time, then ξ_2 is a constant quantity. Then equation (13) is easily integrated:

$$w(t,z)=2B\langle{}^1/_2|\mu_z|{}^1/_2\rangle + {}^1/_2\xi_2\langle{}^1/_2|T_{\Gamma_4 0}|{}^1/_2\rangle\tilde{I}(t,z),$$
$$\tilde{I}(t,z)=\frac{1}{T_1}\int_{-\infty}^{t} I(t',z)\exp\left(\frac{t'-t}{T_1}\right)dt'. \qquad (15)$$

Since $I(t,z)$ actually depends only on the single variable η, it is easily shown that the quantities $\tilde{I}$ and w are also functions of the single variable η. Therefore equations (14) can be integrated in the general form

$$\xi_1(t,z)=f_1(\eta)\cos[aw(\eta)\zeta]+f_2(\eta)\sin[aw(\eta)\zeta],$$
$$\xi_3(t,z)=-f_2(\eta)\cos[aw(\eta)\zeta]+f_1(\eta)\sin[aw(\eta)\zeta], \qquad (16)$$

where $f_{1,2}$ are arbitrary functions. If we take the values of the Stokes parameters to be constant in time on the forward plane of the crystal $z=0$, then these functions must be chosen as follows:

$$f_1(\eta)=\xi_1^{(0)}\cos[aw(\eta)\eta]+\xi_3^{(0)}\sin[aw(\eta)\eta],$$
$$f_2(\eta)=\xi_1^{(0)}\sin[aw(\eta)\eta]-\xi_3^{(0)}\cos[aw(\eta)\eta].$$

Substituting these expressions in (16), we get

$$\xi_1(t,z)=\xi_1^{(0)}\cos 2\varphi-\xi_3^{(0)}\sin 2\varphi,$$
$$\xi_3(t,z)=\xi_3^{(0)}\cos 2\varphi+\xi_1^{(0)}\sin 2\varphi, \qquad (17)$$
$$\varphi=(akz/\omega)w(\eta).$$

The angle φ in (17) determines the rotation of the axes of the polarization ellipse in the plane z at the instant t with respect to the axes of the ellipse in the plane $z=0$. With neglect of effects that are nonlinear in the radiation intensity, the angle φ determines the ordinary Faraday rotation, characterized by the paramagnetic Verdet constant V:

$$\varphi=VBz, \quad V=(2\pi kN/\Theta\varepsilon_0)\langle{}^1/_2|\mu_z|{}^1/_2\rangle\langle{}^1/_2|T_{\Gamma 40}|{}^1/_2\rangle. \qquad (18)$$

The nonlinear autorotation of the polarization ellipse is determined by the second term in the formula (15) for $w(\eta)$.[2)] In the present case the effect has an inertial character, since it is due to the splitting of the doublet sublevels in a variable field and to the consequent change of their populations as a result of relaxation processes.

For pulses of long duration, $\tau\gg T_1$, the form of the function $\tilde{I}(\eta)$ practically coincides with the envelope of the pulse $I(\eta)$. This fact has a simple physical significance: for fast relaxation processes, the ionic subsystem succeeds in readjusting itself to the instantaneous value of the intensity. In the contrary limiting case, $\tau\ll T_1$, the relaxation processes are unable to operate, and the nonlinear autorotation is small.

For Gaussian intensity envelope $I(\eta)=I_0\exp(-\eta^2/\tau^2)$, we have for the function $\tilde{I}$

$$\tilde{I}(\eta)=(\pi^{1/2}\tau/2T_1)\exp\{-(\tau/2T_1)^2-\eta/T_1\}\{1+\Phi(\eta/\tau-\tau/2T_1)\}I_0, \qquad (19)$$

where Φ is the error integral. Figure 1 shows the function (19) for several values of the parameter τ/T_1.

We shall give numerical estimates of the nonlinear autorotation. For $\tau\gg T_1$, $\xi_2\sim 1$, $E_0\sim 5\cdot 10^4$ V/cm, $N\approx 10^{18}$ cm^{-3}, $T\sim 10$ K, and a mistuning of the frequency ω with respect to the ion absorption line ~100 cm^{-1}, we get an angle of rotation of a few radians for a crystal of thickness ~1 cm.

Typical crystals of the type considered in this section are: $CaF_2:Yb^{3+}$, Er^{3+}; $CeO_2:Ho^{2+}$; and others.

6. THE QUASISTATIONARY REGIME

When the inequalities

$$\tau\gg T_{1,2} \qquad (20)$$

are satisfied, the values of the matrix elements of the density matrix follow after the change of radiation in-

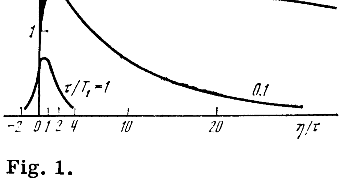

Fig. 1.

tensity adiabatically. In this case we get from equations (7) and (8)

$$u=\lambda I\xi_3,\quad v=\varkappa I\xi_1,\quad w=\Delta,\quad \sigma=\sigma_0. \tag{21}$$

Substituting (21) in (12), we get

$$\begin{gathered}
\frac{\partial\xi_1}{\partial\zeta}=-\frac{\omega}{k}VB\xi_3-\frac{4\pi N\omega}{Q\Theta\varepsilon_0}\nu_Q\xi_2\xi_3I(\beta^{(+)2}+\beta^{(-)2}-\lambda^2),\\
\frac{\partial\xi_2}{\partial\zeta}=\frac{4\pi N\omega}{Q\Theta\varepsilon_0}\nu_Q\xi_1\xi_3I(\varkappa^2-\lambda^2),\\
\frac{\partial\xi_3}{\partial\zeta}=\frac{\omega}{k}VB\xi_1-\frac{4\pi N\omega}{Q\Theta\varepsilon_0}\nu_Q\xi_1\xi_2I(\varkappa^2-\beta^{(+)2}-\beta^{(-)2}),\\
V=\frac{4\pi Nk}{Q\Theta\varepsilon_0}\nu_Q(\beta^{(+)}\delta^{(+)}+\beta^{(-)}\delta^{(-)}),\\
\delta^{(\pm)}=\langle m|\mu_z|m\rangle\mp\langle n|\mu_z|n\rangle,
\end{gathered} \tag{22}$$

where $\nu_Q=1$ for doublets and triplets, $\nu_Q=2$ for quadruplets.

The quantity V is the paramagnetic Verdet constant, which determines the Faraday effect for a weak wave. In the derivation of (22) it has been taken into account that the matrix elements of the operator $T_{\Gamma_3\theta}$ cancel each other for triplets and quadruplets, and that for the doublets Γ_3 they are multipled by the vanishing coefficient $\beta^{(+)}$.

We shall consider specific cases of equations (22).

The doublet Γ_3. In this case $\beta^{(\pm)}=\varkappa=0$. Since then $V=0$, the ordinary Faraday effect is absent in such crystals. We then find from the last equation (22) that ξ_3 can depend only on $\eta=t-kz/\omega$. If the parameter ξ_3 is constant in the incident wave, then it remains constant also in the crystal. This fact is a consequence of the fact that the principal axes of the tensor ε_{ij} coincide in the present case with the crystal axes.

The solution of the first two equations (22) when $\xi_3=\text{const}$ can be written in the form, analogous to (17),

$$\begin{gathered}
\xi_1(z,t)=\xi_1^{(0)}\cos 2\varphi+\xi_2^{(0)}\sin 2\varphi,\\
\xi_2(z,t)=\xi_2^{(0)}\cos 2\varphi-\xi_1^{(0)}\sin 2\varphi,\\
\varphi=\frac{2\pi Nk}{\Theta\varepsilon_0}\xi_3\lambda^2I(\eta)z.
\end{gathered} \tag{23}$$

As is evident, the parameters ξ_1 and ξ_2 are oscillatory functions of the distance of the forward plane of the crystal, varying within the limits $\xi_1^{(0)}+\xi_2^{(0)}$. The period of the oscillations depends on the value of ξ_3. In particular, when $\xi_3=0$ the oscillations are absent, and the polarization ellipse is not deformed during propagation of the wave. It is easily shown that such a solution for $\xi_3=0$ exists for an arbitrary ratio between τ and $T_{1,2}$, and not only when the inequalities (20) are satisfied.

An example of a crystal of this type is $CaF_2:Dy^{2+}$.

Triplet. For triplet states, $\beta^{(-)}=0$. The perturbation of the ionic state is strongest if the frequency of the wave is close to the frequency of the dipole transition of the ion. In this, the most interesting case, one can use for the operators $T_{\Gamma M}$ the single-level approximations; that is, one can neglect in the sum (2) the contribution of other excited states than the resonance state. By using the explicit expressions for the operators $T_{\Gamma M}$, one can show[13] that in the single-level approximation, for the triplet states under consideration the parameters $\beta^{(+)}$, $\varkappa$, and λ are equal in absolute value, provided the resonance state is not the doublet Γ_3. In the contrary case, $\beta^{(+)2}=\varkappa^2=\frac{1}{4}\lambda^2$. In the first case, effects nonlinear in the radiation intensity disappear from equation (22). Change of polarization occurs only in consequence of the ordinary Faraday rotation.

But if the resonance state is the doublet Γ_3, then in the absence of an external magnetic field **B** the parameter ξ_3 is constant. The variation of the parameters ξ_1 and ξ_2 is described by formulas (23) with λ^2 replaced by $\frac{3}{4}\lambda^2$. In the presence of a magnetic field, the solution of equations (22) becomes considerably more complicated. One can find two integrals of the system (22) by dividing the first and third equations by the second:

$$\xi_1^2+\xi_2^2-\frac{2VB\Theta\varepsilon_0}{\pi kN\lambda^2I}\xi_2=C_1,\qquad \xi_3^2+\frac{2VB\Theta\varepsilon_0}{\pi kN\lambda^2I}\xi_2=C_2. \tag{24}$$

Here C_1 and C_2 are constants, related by the condition of complete polarization of the radiation: $C_1+C_2=1$.

Substitution of the integrals (24) in the second equation (22) makes it possible to express ξ_2 in terms of elliptic functions. Complete analysis of the solution, however, requires cumbersome calculations and will not be given here. Examples of crystals with triplet ions: $MgO:Fe^{2+}$, $NaF:Fe^{2+}$.

Quadruplet. Equations (22) simplify in the single-level approximation if the resonance state is a Kramers doublet. In this case, it can be shown[13] that $\lambda^2=\varkappa^2=\frac{4}{3}\beta^{(+)2}=\frac{16}{3}\beta^{(-)2}$. Then from the second equation (22) follows conservation of the parameter ξ_2, which determines the degree of circular polarization of the radiation. The remaining equations for ξ_1 and ξ_3 are easily integrated. The result has the form of formula (17), but now

$$\varphi=VBz-\frac{\pi Nk}{8\Theta\varepsilon_0}\lambda^2I\xi_2z. \tag{25}$$

As is evident, the angle of rotation of the polarization ellipse in this case is the sum of the magnetic rotation and a nonlinear autorotation.

But if in the sum (2) the contribution of intermediate quadruplet states is substantial, equation (22) can not be integrated in terms of elementary functions, since in this case no simple relations can be established between the parameters that occur in the equations. By a procedure similar to the preceding one, one can construct two conserved integrals of the system (22):

$$\begin{gathered}
(\varkappa^2-\lambda^2)\xi_1^2+(\beta^{(+)2}+\beta^{(-)2}-\lambda^2)\xi_2^2+\frac{VB\Theta\varepsilon_0}{\pi kNI}\xi_2=C_1,\\
(\varkappa^2-\lambda^2)\xi_3^2+(\varkappa^2-\beta^{(+)2}-\beta^{(-)2})\xi_2^2-\frac{VB\Theta\varepsilon_0}{\pi kNI}\xi_2=C_2,\\
C_1+C_2=\varkappa^2-\lambda^2.
\end{gathered} \tag{26}$$

By means of the integrals (26), the Stokes parameters in this case can also be expressed in terms of elliptic functions, but the result has a cumbersome form and will not be given here.

Examples of crystals of this type: $CaF_2:Pr^{2+}$, $CaF_2:Dy^{3+}$. The numerical estimates of the nonlinear effects considered in this section coincide with the estimates given in the preceding section for Kramers doublets.

7. THE INVERSE FARADAY EFFECT

The action of a light wave on the paramagnetic ions leads to a change of the populations of the magnetic sublevels, and consequently the magnetization of the crystal changes (the IFE[1]). By definition,

$$\mathbf{M}=NSp(\rho\boldsymbol{\mu}).$$

Since in the Hamiltonian (1), for the chosen directions of the constant magnetic field and of the wave vector of the wave, those operators are absent that produce the harmonics Γ_{4x} and Γ_{4y} corresponding to the operators μ_x and μ_y, the projections of the magnetization along the X and Y axes do not appear. To calculate the magnetization along the z axis, we shall use the formulas obtained earlier for $\rho(z,t)$.

Kramers doublet. In this case

$$M_z=\frac{N}{2\Theta}\left\langle\frac{1}{2}\right|\mu_z\left|\frac{1}{2}\right\rangle w,$$

where the value of w is determined in (13). Using formula (15), we find

$$M_z=\chi B+\frac{\varepsilon_0}{8\pi k}V\xi_2\bar{I}, \tag{27}$$

where $\chi=(N/\Theta)\langle\frac{1}{2}|\mu_z|\frac{1}{2}\rangle^2$ is the paramagnetic susceptibility and where V is the Verdet constant (18). The second term in the expression (27) dexcribes the IFE and reduces to the result of Ref. 1 when $\tau\gg T_1$; then $\bar{I}\approx I$.

Triplets and quadruplets. For the doublets Γ_3, no magnetization occurs either under the action of a constant field $\mathbf{B}$ or under the action of radiation: "nonmagnetic" doublets. For triplets and quadruplets one easily obtains

$$M_z=\frac{N}{2Q\Theta}(\delta^{(+)}w+\delta^{(-)}\sigma), \tag{28}$$

where in the case of quadruplets, $w=w_{3/2}+w_{1/2}$.

In the quasistationary regime, one can show, by using the expressions obtained in the preceding section, that the magnetization is described by formula (27) with $\bar{I}(z,t)$ replaced by $I(z,t)$. Then the paramagnetic susceptibility has the form

$$\chi=\frac{N\nu_Q}{2Q\Theta}(\delta^{(+)2}+\delta^{(-)2}),$$

and the Verdet constant is determined by (22). Although in form formula (27) will agree with the result obtained in Ref. 1, here there is actually an important difference, for now the parameter ξ_2 in general depends on the coordinate z. Therefore in the general case the magnetization induced by the radiation is not constant along the trajectory of the light beam.

8. SHORT LIGHT PULSE IN THE PRESCRIBED-FIELD APPROXIMATION

In order to depart from the framework of the quasistationary approximation, and in particular in order to elucidate the role of combinational scattering (CS) of light, we shall consider the limiting case of a short light pulse:

$$\tau\ll T_{1,2},\ 1/B\delta^{(+)}. \tag{29}$$

The inequalities (29) mean that the spectrum of the incident radiation considerably exceeds both the relaxational width of the magnetic sublevels and their splitting in the constant magnetic field; therefore, evidently, only forced CS is involved.

When (29) is satisfied, the relaxation terms in Equations (7) may be neglected. To eliminate quantities containing the field B, we make the transformation

$$u'=u\cos x+v\sin x,\quad v'=v\cos x-u\sin x,\quad x=B\delta^{(+)}\eta,$$

by means of which equations (7) can be written in the form

$$\begin{gathered}\partial u'/\partial t=(\varkappa\xi_1\cos x-\lambda\xi_3\sin x)Iw-(\xi_2\beta^{(+)}+\gamma)Iv',\\ \partial v'/\partial t=-(\lambda\xi_3\cos x+\varkappa\xi_1\sin x)Iw+(\xi_2\beta^{(+)}+\gamma)Iu',\\ \partial w/\partial t=\lambda\xi_3I(v'\cos x+u'\sin x)-\varkappa\xi_1I(u'\cos x-v'\sin x),\end{gathered} \tag{30}$$

where

$$\gamma=-\frac{1}{4\sqrt{3}}(\langle m|T_{\Gamma_3\theta}|m\rangle-\langle n|T_{\Gamma_3\theta}|n\rangle),$$

for triplets, $\gamma=0$.

Since the intensity $I(\eta)$ is nonzero only within a small neighborhood of the point $\eta=0$, by virtue of the inequalities (29) x may be set equal to zero in equations (30). Furthermore, we shall suppose that the concentration of paramagnetic ions is not too large, or that the crystal thickness is small, so that the variation of the Stokes parameters may be neglected (the prescribed-field approximation). Introducing instead of t a new variable proportional to the intensity area

$$J=\{(\varkappa\xi_1)^2+(\lambda\xi_3)^2+(\beta^{(+)}\xi_2+\gamma)^2\}^{1/2}\int_{-\infty}^{\eta}I(\eta')d\eta', \tag{31}$$

we write equations (30) in the following form:

$$\begin{gathered}\partial\mathbf{R}/\partial J=[\mathbf{SR}],\quad \mathbf{R}=(u',v',w),\\ \mathbf{S}=\{(\varkappa\xi_1)^2+(\lambda\xi_3)^2+(\beta^{(+)}\xi_2+\gamma)^2\}^{-1/2}(\lambda\xi_3,\ \varkappa\xi_1,\ \beta^{(+)}\xi_2+\gamma).\end{gathered} \tag{32}$$

Since equations (32) are equations with constant coefficients, the solutions can be easily found. Supposing that before arrival of the wave $u=v=0$ and $w=B\delta^{(+)}$, we get

$$\begin{gathered}u'=[S_1S_3(1-\cos J)+S_2\sin J]B\delta^{(+)},\\ v'=[S_2S_3(1-\cos J)-S_1\sin J]B\delta^{(+)},\\ w=[S_3^2+(1-S_3^2)\cos J]B\delta^{(+)},\end{gathered} \tag{33}$$

where $S_{1,2,3}$ are the components of the unit vector $\mathbf{S}$ defined in (32). It is obvious that after passage of the pulse the values of u' and v' decay exponentially with relaxation time T_2, while w approaches its equilibrium value in the absence of radiation with relaxation time T_1.

In the short-pulse approximation, the integral of equation (8) is found trivially: σ does not succeed, during the time of the pulse, in changing from its initial value

$$\sigma=2B(\langle 3/2|\mu_z|3/2\rangle+\langle -1/2|\mu_z|-1/2\rangle). \tag{34}$$

The physical meaning of this result is clear; for CS, which, if (29) is satisfied, is solely responsible for change of the populations of the sublevels, is absent for transitions between the quadruplet level pairs $(3/2,-\frac{1}{2})$ and $(\frac{1}{2},-3/2)$.

We shall use the relations obtained to calculate the magnetization of the crystal after passage of the pulse. On substituting (33) and (34) in (28), we get the following results.

For doublets $\Gamma_3, M_z = 0$.

For triplets,

$$M_z = \frac{NB\delta^{(+)2}}{6\Theta} \frac{(\beta^{(+)}\xi_2)^2 + [(\varkappa\xi_1)^2 + (\lambda\xi_3)^2]\cos J}{(\varkappa\xi_1)^2 + (\lambda\xi_3)^2 + (\beta^{(+)}\xi_2)^2}.$$

For quadruplets,

$$M_z = \frac{NB\delta^{(+)2}}{8\Theta}[g^{(+)} + g^{(-)} + (1-g^{(+)})\cos J^{(+)} - (1-g^{(-)})\cos J^{(-)}], \qquad (35)$$

where

$$g^{(\pm)} = \frac{(\beta^{(+)}\xi_2 \pm \lambda/3^{1/2})^2}{(\varkappa\xi_1)^2 + (\lambda\xi_3)^2 + (\beta^{(+)}\xi_2 \pm \lambda/3^{1/2})^2},$$

and where the values $J^{(\pm)}$ are determined by formula (31) with the parameter γ in the square root replaced by $\pm\lambda\ 3^{1/2}$ respectively. This results follows from the equality $\gamma = \pm\lambda\ 3^{1/2}$ for the quadruplet states Γ_8 when $m = 3\ 2$ and $\frac{1}{2}$, which can be obtained after simple calculations.

We shall discuss the formulas (35). For triplets, the change of magnetization is greatest when the intensity area takes the values

$$J = p\pi,\ p = 1, 3, 5\dots. \qquad (36)$$

But if in formula (36) $p = 2, 4, 6, \dots$, the magnetization remains constant. This result is analogous to the well-known one from the theory of self-induced transparency, when for a certain pulse area the system changes completely to the upper one of the resonation states or remains in the lower state.[14] The difference consists of the fact that instead of the pulse area, (36) contains the intensity area. This is due to the fact that in the present case the basic process is a two-photon process, CS, and not single-photon absorption and radiation as for self-induced transparency. It is also noteworthy that the greatest change of magnetization occurs for linearly polarized radiation ($\xi_2 = 0$), when after passage of the pulse the magnetization is changed in direction but remains the same in absolute value as before incidence of the radiation. Furthermore, for completely circularly polarized radiation the crystal magnetization, in the approximation considered, does not change, since then no CS occurs. Here lies the basic difference of the magnetization mechanism determined by CS as compared with the stationary IFE, which is due solely to the circular polarization of part of the radiation.

Similar results follow also for the magnetization of crystals with ions in quadruplet states. But in this case it is impossible to obtain a change of sign of the magnetization without change of its value.

We shall consider a numerical example. In fields $B \sim 1$ kOe, magnetization reversal of the crystal is possible for $E_0 \sim 3 \cdot 10^4$ V/cm, tuning out of the resonance frequency $\sim 10^2$ cm^{-1}, $\tau \sim 10^{-9}$ sec. The change of magnetization by a short light pulse may prove useful for measurement of short relaxation times, and also in optoelectronics.

TABLE I.

SM''	JM'	SM	$\left\langle \begin{matrix} S & J & S \\ M'' & M' & M \end{matrix} \right\rangle$	SM''	JM'	SM	$\left\langle \begin{matrix} S & J & S \\ M'' & M' & M \end{matrix} \right\rangle$
SM	00	$S\,{-M}$	$(-1)^{S+M}/\sqrt{Q}$	2ε	20	2ε	$^1/_2$
$1M$	20	$1\,{-M}$	$(2-\lvert M\rvert)/2\sqrt{3}$	$^1/_2\,M$	30	$^1/_2\,{-M}$	$1/\sqrt{6}$
$1M$	2ε	$1M$	$\lvert M\rvert/2$	$^3/_2\,M$	20	$^3/_2\,{-M}$	$-M/(\lvert M\rvert\sqrt{8})$
$1M$	30	$1\,{-M}$	$-M/\sqrt{6}$	$^3/_2\,M$	2ε	$^3/_2\,3M$	$M/\sqrt{2}$
$1M$	40	$1M$	$M/\sqrt{6}$	$^3/_2\,M$	40	$^3/_2\,3M$	$^1/_2\sqrt{3}$
2θ	20	2θ	$-^1/_2$				

APPENDIX

In this paper, the basis chosen for the unperturbed states of the ion is a basis of characteristic functions of the effective spin resulting from the presence of a constant magnetic field. Because of the definite symmetry of the crystal, there occurs a mutual dependence of the matrix elements of the operators $T_{\Gamma M}$, connected with the Wigner-Eckart theorem for point groups. In the literature (see, for example, Ref. 15, where an extensive bibliography is also given), the Wigner coefficients are given for a real basis that differs from that used in the present paper. Therefore we shall briefly formulate a rule for calculation of the matrix elements $T_{\Gamma M}$ in the basis $|\mathrm{S}m\rangle$.

We shall use for $T_{\Gamma M}$ the notation proposed in Ref. 15: $\Gamma_1 \equiv 0$, $\Gamma_3 \equiv 2$, $\Gamma_4 \equiv 3$, $\Gamma_5 \equiv 4$. Thus we have five different operators: $T_0, T_{2\theta}, T_{2\varepsilon}, T_{30}, T_{40}$. The magnetic moment operator realizes the z component of the irreducible representation Γ_4 and therefore possesses the symmetry properties of the operator T_{30}.

We write the Wigner-Eckart theorem

$$\langle Sm|T_{Jm'}|Sm''\rangle = (-1)^{S+J+m} \left\langle \begin{matrix} S & J & S \\ m'' & m' & -m \end{matrix} \right\rangle T_{JS}, \qquad (\mathrm{A.1})$$

where $\langle\dots\rangle$ in the right side is a Wigner coefficient of the point group, and where T_{JS} is a reduced matrix element. One can carry out the calculations of a Wigner coefficient by expansion of the harmonics of the irreducible representations of the symmetry group of the crystal in spherical harmonics, as was proposed in Ref. 15, or by representing the operators T_{Jm} in terms of components of the effective spin. For the doublet Γ_3, it is customary in (A.1) to set $\theta = -\theta, \varepsilon = -\varepsilon$, $(-1)^\theta = (-1)^\varepsilon = 1$.

Table I gives the nonvanishing Wigner coefficients for those irreducible representations that were considered in the present paper. The other Wigner coefficients are obtained from those in the table by permutation of any two columns. For an odd permutation, the coefficient is multiplied by a factor $(-1)^{2S+J}$.

For the operator T_{30}, odd with respect to time reversal, the relation (A.1) is not satisfied within the quadruplet Γ_8, since the symmetrized product $\{\Gamma_8 \times \Gamma_8\}$ contains the representation Γ_4 twice. But for us, the property of the matrix elements of T_{30} that follows from the Kramers theorem is sufficient: $\langle \frac{3}{2} - m | T_{30} | \frac{3}{2} - m\rangle = -\langle \frac{3}{2} m | T_{30} | \frac{3}{2} m\rangle$. The operator T_{30} has no nondiagonal matrix elements.

The operator T_{40} is even with respect to time reversal; and since the antisymmetrized product $[\Gamma_8 \times \Gamma_8]$

contains the representation Γ_5 once, (A.1) is satisfied for T_{40}. From the table it is evident that $\langle \frac{3}{2}m | T_{40} | \frac{3}{2}n \rangle = -\langle \frac{3}{2}n | T_{40} | \frac{3}{2}m \rangle$; therefore the matrix elements of this operator are pure imaginary.

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[1] The saturation range, however, is not considered in this paper.

[2] A large number of mechanisms of nonlinear autorotation of the polarization ellipse have been discussed earlier for various media (see, for example, Ref. 11 and references cited in that paper, and also Ref. 12). But the treatment was everywhere limited to stationary fields.

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Polarization of characteristic x rays excited by proton impact

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The degree of polarization of the x-ray lines L_l, $L_{\alpha 1,2}$, and $L_{\beta 2,15}$ of a proton-excited silver atom was measured. It is established that the measured degree of polarization of the L_l line decreases from 29 to 8% when the proton energy is increased from 150 to 500 keV. The degree of polarization of the radiation of the investigated lines, calculated in the Born approximation with allowance for the Koster-Kronig transitions, agrees well with experiment. The effect of the polarization on the measurements of the cross sections of the generated x-ray lines is analyzed.

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1. INTRODUCTION

Excitation of atoms by a directional beam of particles produces an aligned state, and the light emitted in the course of its decay is linearly polarized. However, only relatively recently[1] was it understood that the an ion with a vacancy in the inner shell and with total angular momentum $j \geqslant 3/2$, produced when the atom is ionized by electron or proton impact, should also be in an aligned state. This alignment is due to the fact that the cross sections for the ionization are different for states with different values of the modulus of the projection of the angular momentum on the direction of the particle beam. Therefore the x rays accompanying the filling of the vacancies should be anisotropic and polarized. The first successful measurement of the polarization of the characteristic x rays was carried out in Ref. 2, where polarization of the L_{α_1} line of mercury excited by an electron beam was observed.

A theoretical analysis[3,4] has shown that the polarization of the x rays when atoms are ionized by protons can be much larger than the maximum polarization reached in ionization by electrons. A high degree of polarization was predicted in the proton relative velocity region $v/v_0 < 1$ (v_0 is the electron velocity on the given subshell, and v is the velocity of the incident