# Inverse Faraday effect in magnetically ordered crystals

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A study is made of the change in the magnetization of magnetically ordered crystals under the action of laser radiation. The study of this effect in a ferrimagnet yields for the parameters of the magnetic sublattices new information that cannot be obtained from the ordinary magnetooptic effects. The excitation of spin waves by a laser pulse is also studied.

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## 1. INTRODUCTION

Laser radiation with nonzero circular polarization propagating in a medium produces in the latter a magnetization proportional to the radiation intensity. This phenomenon is known as the inverse Faraday effect.[1] This nonlinear magnetooptic effect has been studied theoretically in many papers, including a study of the case of paramagnetic crystals.[2] The present paper is devoted to the inverse Faraday effect in nonconducting magnetically ordered crystals.

As we shall see, the study of the inverse Faraday effect yields valuable information on the structure of magnetic crystals. In addition, the possibility of using the inverse Faraday effect to count single optical photons has been discussed in the literature.[3] We might also mention a study[4] of the absorption of circularly polarized optical radiation in magnetic semiconductors, an absorption that leads to a change in the exchange interaction constant.

A distinguishing feature of magnetically ordered media having a spontaneous or induced magnetization in the absence of radiation is the presence of resonant frequencies corresponding to the excitation of collective fluctuations of the spins. The effective magnetic field due to a pulse of electromagnetic radiation propagating in the medium will therefore excite spin waves whose amplitude is proportional to the intensity of the incident radiation.

Spin waves can be generated by optical radiation by two different mechanisms. The first mechanism involves a change, due to stimulated Raman scattering,[2] in the level populations of the paramagnetic ions contained in the magnetic crystal and decreases the magnetization of the crystal in comparison with its saturated state. The second mechanism involves the disorientation of the magnetization vector at different points in the crystal, with the absolute magnitude of the vector remaining unchanged. Here we shall consider only the second mechanism for the excitation of spin waves. The effect of the first mechanism can be ignored under the condition

$$\Delta\Omega \ll W_{ex}, \tag{1}$$

where $\Delta\Omega$ is the spectral width of the incident radiation and $W_{ex}$ is the energy of the exchange interaction responsible for the level splitting in the paramagnetic ion; in order of magnitude $W_{ex} \simeq T_C$, where $T_C$ is the Curie temperature in energy units.

In the next section we give a phenomenological Hamiltonian for the interaction of a magnetic crystal with optical radiation and find an expression for the effective magnetic field in the crystal. In Sec. 3 we examine the inverse Faraday effect under steady-state conditions, when the radiation intensity is not changing in time. In Sec. 4 we examine the Landau-Lifshitz equations for effects due to the time-varying nature of the radiation. In the subsequent sections we use these equations to study spin waves excited by radiation in a crystal.

## 2. EFFECTIVE MAGNETIC FIELD

Let us suppose we have a crystal of cubic or uniaxial symmetry containing one or two magnetic sublattices. Laser radiation at a frequency far from the absorption lines of the crystal is propagating through it. We average the Hamiltonian $\mathscr{H}$ which is quadratic in the amplitudes of the fields **E** and **H**, over the rapid oscillations of the field, and write it in the form of an expansion in powers of the magnetization **M**, keeping only the linear terms (cf. Refs. 5 and 6):

$$\begin{aligned}\mathscr{H} = {} & \mathscr{H}_0(\mathbf{M}^{(p)}) + \mathscr{H}_1(\mathbf{E},\mathbf{H}) \\ & - i\sum_p \mathbf{M}^{(p)}\{\xi_p^{\,e}[\mathbf{E}\times\mathbf{E}^*] + \xi_p^{\,m}[\mathbf{H}\times\mathbf{H}^*]\} \\ & - i\sum_p (\mathbf{n}\mathbf{M}^{(p)})\{\eta_p^{\,e}([\mathbf{E}\times\mathbf{E}^*]\mathbf{n}) + \eta_p^{\,m}([\mathbf{H}\times\mathbf{H}^*]\mathbf{n})\}\end{aligned} \tag{2}$$

Here $\mathscr{H}_0$ is the Hamiltonian of the crystal in the absence of radiation, $\mathscr{H}_1$ is the interaction Hamiltonian of the crystal with the radiation in the paramagnetic state, which is quadratic in the amplitudes of the fields **E** and **H**, and **n** is the anisotropy axis for a uniaxial crystal. The index $p$ in (2) assumes the value $p = 1$ if there is a single magnetic sublattice, or the values $p = 1, 2$ if there are two such sublattices; $\xi_p^{e,m}$ and $\eta_p^{e,m}$ are magnetooptic coefficients due to the gyroelectric and gyromagnetic properties of the crystal, respectively. For cubic crystals $\eta_p^{e,m} = 0$. Since we are not considering magnetoelectric media, the Hamiltonian (2) does not contain cross terms in the amplitudes of **E** and **H** (Ref. 7).

The effective magnetic field acting on the $p$-th sublattice can be found from (2) as

$$\mathbf{H}^{(p)} = -\delta\mathscr{H}/\delta\mathbf{M}^{(p)} = \mathbf{H}_0^{(p)} + \mathbf{H}_I^{(p)}, \tag{3}$$

where $\mathbf{H}_0^{(p)} = -\delta\mathscr{H}_0/\delta\mathbf{M}^{(p)}$ is the effective magnetic field in the absence of radiation, and

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$$\mathbf{H}_I^{(p)}=i\{\xi_p^e[\mathbf{E}\times\mathbf{E}^*]+\xi_p^m[\mathbf{H}\times\mathbf{H}^*]\}$$
$$+in\{\eta_p^e([\mathbf{E}\times\mathbf{E}^*]\mathbf{n})+\eta_p^m([\mathbf{H}\times\mathbf{H}^*]\mathbf{n})\} \quad (4)$$

is the effective magnetic field due to the inverse Faraday effect. It is seen from (4) that $\mathbf{H}_I^{(p)}=0$ for linearly polarized radiation. This circumstance is due to two approximations. First, as already mentioned, we are ignoring the magnetization change due to stimulated Raman scattering, which is small under condition (1). Second, we have dropped terms from the Hamiltonian (2) that are quadratic in the magnetization, and these would lead to $\mathbf{H}_I^{(p)}\neq 0$ for linearly polarized light (see, for example, Ref. 8, were the case of a magnetoactive gas of free electrons is considered). Estimates show that for magnetic crystals the ratio of the quadratic corrections to the linear terms is of order $|\Delta n_l/\Delta n_c|$, where $\Delta n_{l,c}$ is the refractive-index change responsible for the linear and circular birefringence, respectively. For most crystals this ratio is small.[9]

Let us relate the parameters $\xi_p$ and $\eta_p$ to the quantities which characterize the magnetooptic effects in the linear theory. To do this, we introduce the polarizability tensor $\alpha_{ij}(\omega)$ and the magnetic susceptibility tensor $\chi_{ij}(\omega)$:

$$\alpha_{ij}(\omega)=-\partial^2\mathcal{H}/\partial E_i^*\partial E_j, \quad \chi_{ij}(\omega)=-\partial^2\mathcal{H}/\partial H_i^*\partial H_j.$$

We note that writing the tensors $\alpha_{ij}$ and $\chi_{ij}$ in this form presupposes that the electric and magnetic fields in the wave are of the form

$$\mathbf{E}=\mathbf{E}(\mathbf{r},t)e^{i\mathbf{kr}-i\omega t}+\text{c.c.}, \quad \mathbf{H}=\mathbf{H}(\mathbf{r},t)e^{i\mathbf{kr}-i\omega t}+\text{c.c.}$$

Using expression (2), we find

$$\alpha_{ij}(\omega)=\alpha_{ij}^{(0)}(\omega)-i\sum_p e_{ijk}[\xi_p^e M_k^{(p)}+\eta_p^e n_k(\mathbf{nM}^{(p)})],$$
$$\chi_{ij}(\omega)=\chi_{ij}^{(0)}(\omega)-i\sum_p e_{ijk}[\xi_p^m M_k^{(p)}+\eta_p^m n_k(\mathbf{nM}^{(p)})]. \quad (5)$$

Here $e_{ijk}$ is the antisymmetric unit tensor, and $\alpha_{ij}^{(0)}$ and $\chi_{ij}^{(0)}$ are the polarizability and magnetic susceptibility tensors of the crystal in the paramagnetic state. For optical frequencies one may set $\chi_{ij}^{(0)}=0$. Furthermore, we shall neglect the crystal birefringence due to its uniaxiality in comparison with the birefringence due to its magnetic ordering, i.e., we set

$$\varepsilon_\parallel^0\approx\varepsilon_\perp^0=\varepsilon_0, \quad 4\pi\alpha_{ij}^{(0)}=(\varepsilon_0-1)\delta_{ij},$$

where $\varepsilon_{\perp,\parallel}^0$ are the principal values of the permittivity tensor of the crystal in the paramagnetic state.

Let us consider a ferromagnet with two magnetic sublattices, which are found in the ground state. For simplicity we shall assume that in the case of uniaxial crystals there is anisotropy of the easy-axis type, $\mathbf{M}^{(1)}+\mathbf{M}^{(2)}\perp\mathbf{n}$, or of the easy-plane type, $\mathbf{M}^{(1)}+\mathbf{M}^{(2)}\parallel\mathbf{n}$. For radiation propagating along the direction of the vector $\mathbf{M}^{(1)}$, which we take to be the $z$ axis, the angle of rotation of the plane of polarization per unit length of crystal is given by[5]

$$\Theta(\omega)=\frac{2\pi i\omega n_0}{c}[\alpha_{xy}(\omega)/\varepsilon_0+\chi_{xy}(\omega)], \quad (6)$$

where $n_0+\varepsilon_0^{\frac{1}{2}}$. Substituting expressions (5) into (6) we find for the case of easy-axis anisotropy

$$\Theta(\omega)=K_1(\omega)M^{(1)}-K_2(\omega)M^{(2)}, \quad K_p(\omega)=2\pi\omega(\bar{\xi}_p+\bar{\eta}_p)/cn_0,$$
$$\bar{\xi}_p=\xi_p^e+\varepsilon_0\xi_p^m, \quad \bar{\eta}_p=\eta_p^e+\varepsilon_0\eta_p^m; \quad (7)$$

Here $K_p(\omega)$ is the Kundt constant for the $p$-th sublattice. In the case of easy-plane anisotropy or in the case of cubic crystals, one should set $\bar{\eta}_p=0$ is the expression for $K_p$. The constants $\bar{\xi}$ and $\bar{\eta}$ can thus be related to the Kundt constants of the corresponding lattices, which determine the angle of rotation of the plane of polarization of radiation propagating in a magnetically ordered medium.

With allowance for the relation $\mathbf{H}\times\mathbf{H}=n_0\mathbf{E}\times\mathbf{E}$ and the assumptions made above, expression (4) for the effective magnetic field can be rewritten in the form

$$\mathbf{H}_I^{(p)}=\frac{2\pi A}{cn_0}I\{\mathbf{v}\bar{\xi}_p+\mathbf{n}(\mathbf{vn})\bar{\eta}_p\},$$
$$A\mathbf{v}=\frac{i[\mathbf{EE}^*]}{(\mathbf{EE}^*)}, \quad I=\frac{cn_0}{2\pi}(\mathbf{EE}^*), \quad (8)$$

where $\varkappa_{ij}^{(p)}=\partial\mathbf{M}_i^{(p)}/\partial H_{ej}$ is the differential static susceptibility tensor of the $p$-th sublattice in an external magnetic field $\mathbf{H}_e$, assumed for simplicity to be directed along the axis of the vector $\mathbf{M}^{(1)}$.

## 3. STEADY-STATE CONDITIONS

Let us consider the case of time-independent radiation intensity. If the sublattice magnetizations are not saturated, the effective magnetic field (8) causes a change in the magnetization of the crystal:

$$\Delta M_i=\sum_p \varkappa_{ij}^{(p)}(\mathbf{H}_e)H_{Ij}^{(p)}, \quad (9)$$

where $\varkappa_{ij}^{(p)}=\partial\mathbf{M}_i^{(p)}/\partial H_{ej}$ is the differential static susceptibility tensor of the $p$-th sublattice in an external magnetic field $\mathbf{H}_e$, assumed for simplicity to be directed along the axis of the vector $\mathbf{M}^{(1)}$.

Let us take up in turn the cases of ferro-, ferri-, and antiferromagnetic crystals.

*Ferromagnets.* In this case the summation index $p$ in (9) takes on a single value:

$$\Delta M_i=\frac{2\pi A}{cn_0}I\varkappa_{ij}(\mathbf{H}_e)\{v_j\bar{\xi}+n_j(\mathbf{nv})\bar{\eta}\}. \quad (10)$$

In particular, for the case of cubic crystals ($\varkappa_{ij}=\varkappa\delta_{ij}$) we have

$$\Delta\mathbf{M}=\frac{AK(\omega)}{\omega}\varkappa(\mathbf{H}_e)I\mathbf{v}. \quad (11)$$

Expression (11) is of the same form as the result of Pershan[1] for the inverse Faraday effect in weakly magnetic media.

*Ferrimagnets.* For cubic crystals such as yttrium iron garnet

$$\Delta\mathbf{M}=\frac{AI\mathbf{v}}{\omega}\{\varkappa^{(1)}(\mathbf{H}_e)K_1(\omega)+\varkappa^{(2)}(\mathbf{H}_e)K_2(\omega)\}. \quad (12)$$

Expressions (7) and (12) imply that one cannot describe the inverse Faraday effect in a two-sublattice ferromagnet by using the parameters characterizing the linear properties of the crystal. In fact, while the angle of rotation of the plane of polarization of the radiation is determined by the difference between the contributions of the two sublattices (7), the mag-

netization due to the inverse Faraday effect is determined by the sum of the sublattice contributions. This circumstance can be understood by recognizing that elliptically polarized radiation "twists" the atomic electrons of both sublattices in the same direction, giving rise to a magnetic field of the same sign in the two sublattices, whereas the contributions of the different sublattices to the rotation of the plane of polarization are of opposite sign as a result of the antiparallel orientation of the spin moments.

Thus the study of the inverse Faraday effect in ferrimagnets yields additional information that cannot be extracted from linear magnetooptic experiments. Up till now it had been assumed that the inverse Faraday effect would give no new information about the properties of a medium that could not be obtained from the ordinary Faraday effect.

*Antiferromagnets.* Assuming that for mirror-image sublattices we have

$$\xi_1=\xi_2=\xi, \quad \eta_1=\eta_2=\eta \tag{13}$$

and introducing the total magnetic susceptibility tensor[9] $\varkappa=\varkappa^{(1)}+\varkappa^{(2)}$, one can easily obtain expression (11) for the magnetization of an antiferromagnet. Thus, in regard to the inverse Faraday effect an antiferromagnet is equivalent to a ferromagnet whose parameters $\bar{\xi}$ and $\bar{\eta}$ are determined by one of the mirror-image sublattices and whose tensor $\varkappa$ is the total magnetic susceptibility.

Simple estimates show that for divalent compounds of iron and europium ($FeCl_2$, EuSe), and also for gallium–yttrium iron garnets,[10] which are transparent in the visible and infrared and have Verdet constants $V(\omega)=K(\omega)\varkappa\simeq 1$–10 deg/cm·Oe, the magnetization reaches values of 0.025–0.25 G at $E=10^4$ V/cm.

## 4. THE LANDAU-LIFSHITZ EQUATION

In the case of radiation of varying intensity, we shall assume that the characteristic time $\tau_I$ for changes in the intensity is significantly longer than the correlation time $\tau_c$ of the lattice. This assumption means that the lattice, which plays the role of a constant-temperature reservoir in regard to the magnetization, is rapidly changing its state and assumes an equilibrium value for each new value of the radiation intensity. For typical crystals one has $\tau_c\simeq 10^{-13}$–$10^{-12}$ s. With such a restriction on one can use as before the formulas obtained above, regarding the electric and magnetic field amplitudes as slowly varying in time. The effective magnetic field (8) plays here the role of an external field capable of causing oscillations of the magnetization of the crystal. For such oscillations to be excited it is necessary that $\mathbf{H}_I$ be noncollinear with the equilibrium magnetization vector.

The intensity $I$ contained in expression (8) for the effective magnetic field can be written for the case of time-varying radiation in the form

$$I=I(t-\mathbf{r}\nu/v,\ \mathbf{r}), \tag{14}$$

where $\mathbf{v}=c\nu/n_0$ is the velocity of propagation of the radiation, while the first argument in expression (14) gives the translation of the pulse in space, and the second determines the transverse-intensity profile of the pulse.

The fluctuations of the crystal magnetization are described by the Landau-Lifshitz equations for the respective sublattices. In particular, in the approximation linear in the perturbing field (8) these equations become

$$\begin{gathered}\partial\mathbf{M}_{\sim}^{(p)}/\partial t=-\gamma_p[\mathbf{M}_{\sim}^{(p)}\times\mathbf{H}_0^{(p)}]-\gamma_p[\mathbf{M}_0^{(p)}\times\mathbf{H}_I^{(p)}]+\mathbf{R}_p,\\ \mathbf{M}^{(p)}=\mathbf{M}_0^{(p)}+\mathbf{M}_{\sim}^{(p)},\end{gathered} \tag{15}$$

where $\gamma_p$ is the gyromagnetic ratio, $\mathbf{M}_{\sim}^{(p)}$ and $\mathbf{M}_0^{(p)}$ are the alternating and equilibrium components of the $p$-th sublattice magnetization, $M_{\sim}^{(p)}\ll M_0^{(p)}$, $\mathbf{H}_0^{(p)}$ is given by formula (3), and

$$\mathbf{R}_p=\frac{\beta_p}{M_0^{(p)}}\left[\mathbf{M}_0^{(p)}\times\frac{\partial\mathbf{M}_{\sim}^{(p)}}{\partial t}\right], \tag{16}$$

is a relaxation term in the Gilbert form,[11] with the phenomenological damping parameter $\beta_p$ considered small: $\beta_p\ll 1$.

## 5. HOMOGENEOUS PRECESSION

Let the dimensions $L$ of the sample be much smaller than the characteristic dimensions $a$ of the transverse inhomogeneity of the radiation. The intensity (14) can then be assumed to depend only on the first argument:

$$I=I(t-\mathbf{r}\nu/v).$$

We shall also neglect the contribution from magnetic fields of the type $\mathbf{H}_{\text{inhom}}^{(p)}\|\alpha_p\Delta\mathbf{M}^{(e)}$, which result from the inhomogeneity of the exchange interaction, retaining only the magnetic fields due to the homogeneous intersublattice exchange interaction. This situation corresponds to the excitation of long-wavelength spin waves or of uniform precession of the magnetization, the latter being possible under the following restrictions on the dimensions of the sample[12]: $\alpha^{1/2}\ll L\ll a$. when these conditions are satisfied, the effective magnetic field (3) can be written[11]

$$H_i^{(1,2)}=-\delta M_i^{(2,1)}+H_{ai}^{(1,2)}+H_{ei}-4\pi N_{ij}(M_j^{(1)}+M_j^{(2)})+H_{Ii}^{(1,2)}. \tag{17}$$

Here $\delta$ is the homogeneous exchange interaction constant, $H_a$ is the anisotropy magnetic field, and $N_{ij}$ is the demagnetizing-coefficient sample in the shape of an ellipsoid of revolution with easy-axis anisotropy, the case to which all further discussion will be limited, we have

$$N_{xx}=N_{yy}=N_\perp, \quad N_{zz}=N_3, \quad 2N_\perp+N_3=1,$$

$$\mathbf{H}_a^{(p)}=-2C_pM_z^{(p)}\mathbf{e}_z, \quad C_p<0.$$

Further, since the precession of the magnetization is caused by only the perpendicular component of the alternating field, we can without loss of generality take the radiation propagation direction to be along the $x$ axis. In this case the magnetic field due to the inverse Faraday effect may be written in the form

$$\mathbf{H}_I^{(p)}=\frac{AK_p(\omega)}{\omega}I\left(t-\frac{x}{v}\right)\mathbf{e}_x. \tag{18}$$

Here

$$K_p(\omega)=\frac{2\pi\omega}{cn_0}\bar{\xi}_p.$$

Thus, in the case of radiation propagating perpendicular to the anisotropy axis, only the magnetooptic coefficients $\bar{\xi}_p$

due to the isotropic part of the Hamiltonian of the magnetic crystal contribute to the effective magnetic field.

The excitation of uniform precession of the magnetization by means of the effective magnetic fields (18) cannot be described in a two-sublattice ferrimagnet by the magnet susceptibility tensor $\chi_{ij}$. This circumstance is due to the fact that the amplitudes of the fields (18) acting on each of the sublattices are different, in contrast to the usual situation that arises in the theory of magnetic resonance. Introducing in (15) the variable $\zeta = t - x/v$ and transforming to the circular components of the magnetization

$$M_{\sim x}^{(p)} \pm i M_{\sim y}^{(p)} = M_{\pm}^{(p)},$$

we obtain the following equations for the Fourier components of the magnetization:

$$\begin{aligned}
&[\Omega(1-i\beta_1)+\gamma_1(H_1+4\pi N_\perp M_0^{(1)})]M_+^{(1)} \\
&+\gamma_1[H_{2E}+4\pi N_\perp M_0^{(1)}]M_+^{(2)} = \gamma_1 M_0^{(1)} H_I^{(1)}(\Omega), \\
&-\gamma_2[H_{1E}+4\pi N_\perp M_0^{(1)}]M_+^{(1)} + [\Omega(1+i\beta_2) \\
&+\gamma_2(H_2-4\pi N_\perp M_0^{(2)})]M_+^{(2)} = -\gamma_2 M_0^{(2)} H_I^{(2)}(\Omega),
\end{aligned} \tag{19}$$

$$M_0 = M_0^{(1)} - M_0^{(2)}, \quad H_I^{(p)} = \frac{1}{2\pi}\int_{-\infty}^{\infty} H_I^{(p)}(t) e^{-i\Omega t} dt.$$

Here

$$H_{1,2E} = \delta M_0^{(2,1)}, \quad H_{1,2} = \pm H_{1,2E} \pm H_a^{(1,2)} - 4\pi M_0 N_3 + H_e,$$

where the sign $\pm$ refers to indices 1 and 2. The equations for the component $M_{\pm}^{(p)}$ are obtained from (19) by the replacement

$$\gamma_p \to -\gamma_p, \quad \beta_p \to -\beta_p.$$

Let us again consider the cases of ferro-, ferri-, and antiferromagnetic media.

*Ferromagnets.* In the case of a single sublattice we set $M_{\pm}^{(2)} = 0$ in equations (19) and obtain the following expression for the alternating component of the magnetic moment along the direction of propagation of the radiation:

$$M_x(\zeta) = \frac{AK(\omega)\gamma M_0}{\omega}\int_{-\infty}^{\zeta} I(\zeta') e^{-(\zeta-\zeta')/\tau} \sin\omega_0(\zeta-\zeta')\, d\zeta'. \tag{20}$$

Here $\omega_0 = \gamma[H_e + H_a - 4\pi M_0(N_3 - N_\perp)]$ is the ferromagnetic resonance frequency, and $\tau^{-1} = \beta\omega_0$ is the reciprocal relaxation time for the magnetization. Introducing the variable $s = (\zeta' - \zeta)/\tau$, we rewrite Eq. (20) in the following form:

$$\begin{aligned}
M_x(\zeta) &= \frac{AK(\omega)\gamma M_0 \tau}{\omega} I(\zeta, \omega_0, \tau), \\
I(\zeta, \omega_0, \tau) &= \int_0^{\infty} I(\zeta - \tau s) e^{-s} \sin(\omega_0 \tau s)\, ds.
\end{aligned} \tag{21}$$

If the pulse is of short duration, the function $I(\zeta - \tau s)$ has a sharp peak at $s = \zeta/\tau$ with a width $\Delta s = \tau_I/\tau$. Therefore, under the conditions

$$\omega_0 \tau_I \ll 1, \quad \tau_I/\tau \ll 1 \tag{22}$$

we may take the slowly varying function out from under the integral sign.

We then obtain the following expression for the magnetization:

$$M_x(\zeta) = \frac{AM_0K(\omega)\gamma}{\omega} e^{-\zeta/\tau}\sigma_I(\zeta)\sin(\omega_0\zeta), \tag{23}$$

where

$$\sigma_I(\zeta) = \int_{-\infty}^{\zeta} I(s)\, ds$$

is the area of the intensity envelope of the laser radiation, which can be regarded as the total energy of the radiation passing through a unit area of the crystal at a given point $x$ prior to time $t$.

The most interesting question is the behavior of the magnetization after the laser pulse has passed through the crystal, i.e., at times $\zeta \gg \tau_I$. In this case the upper limit in (23) for the area of the pulse envelope can be replaced by infinity, and the magnetization will thereby be independent of the shape of the pulse, depending only on the total area of the intensity (cf. Ref. 2). It follows from (23) that for excitation by a short laser pulse the magnetization undergoes damped oscillations at the ferromagnetic resonance frequency.

In the other limiting case, when the pulse is long compared to $\tau$, the main contribution to the integral in (21) is from the region $s \simeq 0$. In this case

$$M_x(\zeta) = \frac{AM_0K(\omega)\gamma\tau^2\omega_0}{2\omega(1+\omega_0^2\tau^2)} I(\zeta). \tag{24}$$

This result means that for long light pulses the variable component of the magnetization that is directed along the laser beam adiabatically follows the change in the intensity envelope of the radiation.

*Ferrimagnets.* An analogous treatment can be applied to the case of a two-sublattice ferrimagnet. Solving Eqs. (19) with allowance for (18) and (17) for the effective magnetic fields and performing the inverse Fourier transform, we obtain the following expression for the resultant magnetization along the direction of propagation of the radiation:

$$M_x(t) = M_x^{(l)}(t) + M_x^{(h)}(t), \tag{25}$$

where $M_x^{(l,h)}$ are the low- and high-frequency components of the variable magnetization, where the low-frequency component is described by formulas (20)–(24), which were obtained for ferromagnets, with the following replacements:

$$\begin{aligned}
&\gamma \to \gamma_{\text{eff}} = \frac{M_0^{(1)} - M_0^{(2)}}{M_0^{(1)}/\gamma_1 - M_0^{(2)}/\gamma_2}, \\
&\omega_0 \to \omega_E = \gamma_{\text{eff}}[H_e + H_a^{\text{eff}} - 4\pi(M_0^{(1)} - M_0^{(2)})(N_3 - N_\perp)], \\
&M_0 \to M_0^{(1)} - M_0^{(2)}, \quad I \to I/2, \quad H_a^{\text{eff}} = \frac{M_0^{(1)} H_a^{(1)} + M_0^{(2)} H_a^{(2)}}{M_0^{(1)} - M_0^{(2)}}, \\
&\tau \to \tau_l = \omega_l \frac{\beta_1 M_0^{(1)}/\gamma_1 + \beta_2 M_0^{(2)}/\gamma_2}{M_0^{(1)}/\gamma_1 - M_0^{(2)}/\gamma_2}.
\end{aligned} \tag{26}$$

According to Eqs. (26), in the low-frequency region a ferri-

magnet behaves like an effective ferromagnet, in agreement with the familiar results from the theory of the ferromagnetic resonance.[11]

The high-frequency component $\mathbf{M}^{(h)}$ can be written

$$M_x^{(h)}(\xi)=\frac{A(\gamma_1-\gamma_2)\tau_h M_0^{(1)}M_0^{(2)}[\gamma_1 K_1(\omega)+\gamma_2 K_2(\omega)]}{4\omega(\gamma_2 M_0^{(1)}-\gamma_1 M_0^{(2)})}\times \tilde{I}(\xi,\omega_h,\tau_h), \tag{27}$$

where

$$\tau_h^{-1}=\delta(\gamma_1\beta_1 M_0^{(1)}+\gamma_2\beta_2 M_0^{(2)}),$$

and $\omega_h=\delta(\gamma_2 M_0^{(1)}-\gamma_1 M_0^{(2)})$ is the exchange frequency of the ferrimagnetic resonance.

Formula (27) is studied for the cases of short and long light pulses in a manner analogous to the case of ferromagnets. We note that the high-frequency amplitude turns out to be much smaller than the low-frequency amplitude. This is because the frequency $\omega_h$ of the exchange resonance lies in the infrared region of the spectrum: $\omega_h \sim 10^{12}$–$10^{13}$ $s^{-1}$ (Ref. 9). Therefore, even for picosecond pulses we have the condition $\omega_h\tau_I \gtrsim 1$, and the quantity $\tilde{I}(\xi,\omega_h,\tau_h)$ given by formula (21) will be small on account of the rapid oscillations of the integrand.

Thus, after passage of a laser pulse through a ferrimagnetic crystal, the variable magnetization induced in the crystal is mainly due to the low-frequency branch of the spin fluctuations.

*Antiferromagnets.* The variable component of the magnetic moment for an antiferromagnet with two mirror-image sublattices in the ground state can, upon solution of Eqs. (19), be written in the form

$$M_x(\xi)=\frac{AK(\omega)M_0}{\omega}\left(\frac{H_a}{H_E}\right)^{1/2}\gamma\tau_a\tilde{I}(\xi,\omega_a,\tau_a); \tag{28}$$

here $H_E=\delta M_0$ is the exchange field, $H_a=-2CM_0$,

$$\omega_a=\gamma[H_a(H_a+2H_E)]^{1/2}$$

is the antiferromagnetic resonance frequency, and $K(\omega)$ is the Kundt constant corresponding to one of the mirror-image sublattices of the antiferromagnet. We note that because the antiferromagnetic-resonance frequency lies in the infrared, $\omega_a \sim 10^{11}$–$10^{12}$ $s^{-1}$, the excitation of an appreciable magnetization in an antiferromagnet requires picosecond laser pulses. The amplitude of the magnetization (28) even under the condition $\omega_a\tau_I \ll 1$ is smaller than the corresponding amplitude for a ferromagnetic crystal by the factor $(H_a/H_E)^{1/2}\simeq 0.1$.

Let us make some numerical estimates. For $FeCl_2$, EuSe, and gallium–yttrium iron garnet at a pulse length $\tau_I \simeq 10$ ns and a degree of circular polarization $A=1$, the amplitude reaches 1–5 G for a field amplitude in the wave $E=10^4$ V/cm.

Figure 1 shows time dependence of the variable magnetization of a ferromagnetic crystal for the case of a Gaussian intensity envelope $I(\xi)=I_0\exp(-\xi^2/\tau_I^2)$ for two different pulse lengths. It is seen in the figure that an increase in the parameter $\omega_0\tau_I$ leads to a decrease in the number of oscillations of the magnetization after the laser pulse has passed through the crystal.

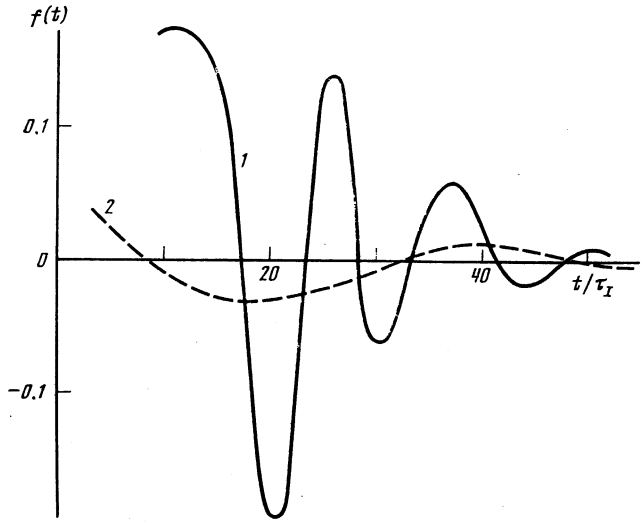


FIG. 1. The time dependence of the homogeneous magnetization of a ferromagnetic crystal: $f(t)=(\tau_I/\tau)\tilde{I}(t)/I_0$. Curve 1) $\omega_0\tau_I=0.5$, $\tau_I/\tau=0.1$, curve 2) $\omega_0\tau_I=1.5$, $\tau_I/\tau=0.01$.

## 6. EXCITATION OF SPIN WAVES

In the preceding sections we have dealt with the excitation of uniform precession of the magnetization by spatially homogeneous laser radiation. Spatially homogeneous electromagnetic fields in the optical region can as well excite spin waves in thin samples and also inhomogeneous Walker fluctuations, by a mechanism analogous to the linear (in the external field) mechanisms for the excitation of spin waves by microwave radiation.[11,12]

The special features of the excitation of spin waves by laser radiation are due to the possibility of focusing the radiation in a relatively small region. Let us consider the excitation of spin waves by such a mechanism for the particular case of a uniaxial ferromagnet. We shall assume that the radiation intensity in the transverse direction has a Gaussian profile:

$$I(\mathbf{r},t)=I(t-x/v)\exp[-(y^2+z^2)/a^2], \tag{29}$$

where $a$ is the width of the Gaussian beam. The effective magnetic field (3) for a uniaxial ferromagnet with allowance for inhomogeneous exchange interactions is of the form[11]

$$H_i=(H_e-4\pi N_3 M_0+H_a)\delta_{iz}+\alpha_1(\partial^2 M_i/\partial x^2+\partial^2 M_i/\partial y^2)+\alpha_2\partial^2 M_i/\partial z^2+h_i(\mathbf{r},t)+H_{Ii}(\mathbf{r},t); \tag{30}$$

here $z$ is the anisotropy axis, $\alpha_{1,2}$ are the inhomogeneous exchange interaction constants, and $\mathbf{h}(\mathbf{r},t)$ is the variable magnetic field due to the spin wave; the Fourier component of this field is given by the relation[12]

$$\mathbf{h}(\mathbf{r},t)=\int d\mathbf{q}d\Omega\mathbf{h}(\mathbf{q},\Omega)\exp(i\mathbf{q}\mathbf{r}-i\Omega t),$$
$$\mathbf{h}(\mathbf{q},\Omega)=-4\pi\mathbf{q}(\mathbf{q}\mathbf{M}(\mathbf{q},\Omega)/q^2). \tag{31}$$

Substituting expressions (29)–(31) into Eq. (15), we find the

following expression for the Fourier component of the magnetization:

$$M_x(\mathbf{q},\Omega)=-\frac{2\pi^2a^2i\gamma^2M_0K(\omega)I(\Omega)\delta(q_z-\Omega/v)}{\Omega_s^2(\mathbf{q})-2i\Omega\nu_s(\mathbf{q})-\Omega^2}\times\frac{H_q\exp(-q_\perp^2a^2/4)}{\omega} \tag{32}$$

Here $\Omega_s(\mathbf{q})=\gamma[H_q(H_q+4\pi M_0\sin^2\theta_q)]^{1/2}$ is the eigenfrequency of the spin fluctuations,

$$H_q=H_e-4\pi N_3M_0+H_a+M_0(\alpha_1\sin^2\theta_q+\alpha_2\cos^2\theta_q)q^2,$$

$\nu_s(\mathbf{q})=\beta\gamma[H_q+2\pi M_0\sin^2\theta_q]$ is the exponential damping constant for the spin wave, and $\theta_q$ is the angle between the wave vector $\mathbf{q}$ and the $z$ axis.

We note now that the intensity envelope $I(\Omega)$ has its maximum at $\Omega\simeq 0$. It follows then from (32) that the resonant excitation amplitude for a spin wave of frequency $\Omega=\Omega_s(\mathbf{q})$ reaches its maximum under the conditions

$$\nu_s(\mathbf{q})\ll\Omega_s(\mathbf{q})\ll\Delta\Omega$$

($\Delta\Omega$ is the spectral width of the laser radiation). It is also seen from (32) that when laser radiation is focused in the interior of a crystal, a beam of spin waves will be excited in the crystal, with a momentum spread $\Delta q_\perp\simeq 2/a$, in the transverse direction. In the direction of propagation of the radiation, on the other hand, the wave vector of the spin wave lies within the interval $q_x\lesssim\Delta\Omega/v$. when the relation

$$n_0a\Delta\Omega/2c\ll 1 \tag{33}$$

is satisfied, the focused laser pulse will excite spin waves in the direction perpendicular to its propagation direction. Assuming for purposes of estimation that $a\simeq 10^{-2}$ cm, we see that inequality (33) is satisfied for pulses with a spectral width $\Delta\Omega\ll 10\ \text{cm}^{-1}$.

## 7. CONDITIONS OF MODULATED INTENSITY

In the previous sections we have considered the excitation of spin waves by single laser pulses. It is clear that the efficiency of excitation of magnetization fluctuations can be increased significantly by having the pulse repetition frequency of the laser coincide with the ferromagnetic resonance frequency, in analogy with the excitation of acoustic fluctuations in liquids by radiation modulated at an acoustic frequency.[13]

In accordance with the foregoing, we write the radiation intensity in the form

$$I(\mathbf{r},t)=I_0\exp[-(y^2+z^2)/a^2](1+m\cos\omega_mt), \tag{34}$$

where $0\leqslant m\leqslant 1$ is the modulation index and $\omega_m$ is the modulation frequency. Using (30), (31), and (34), we obtain from (15) the expression

$$M_x(\mathbf{q},t)=\frac{AK(\omega)M_0\pi a^2\gamma^2H_qm\exp(-q_\perp^2a^2/4)\exp(-i\omega_mt)}{\omega[\Omega_s^2(\mathbf{q})-2i\omega_m\nu_s(\mathbf{q})-\omega_m^2]}. \tag{35}$$

In this case the equality $\omega_m=\Omega_s(q)$ determines the spin-wave numbers at which resonant excitation of the magnetization can occur. All the estimates given for the wave numbers in the previous section remain valid for this case as well. For $q_\perp a\ll 1$, homogeneous magnetization fluctuations

$$M_x(t)=\frac{AK(\omega)M_0mI_0\gamma\omega_0}{\omega}\operatorname{Re}\frac{\exp(-i\omega_mt)}{\omega_0^2-2i\omega_m\tau^{-1}-\omega_m^2}$$

will be excited in the sample, where $\omega_0$ and $\tau$ are determined by relations (20).

For example, for the crystals mentioned in the previous sections, EuSe, $FeCl_2$, and gallium-yttrium iron garnet, the excitation amplitude reaches a value of the order of 25 G for $E=10^4$ V/cm, $\tau=10^{-7}$ s, $K=0.1$ deg/cm·Oe, $M_0\simeq 2\cdot 10^2$ G, $\omega=10^{14}\ \text{s}^{-1}$, and $A=1$.

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