Nonlinear magnetoelectric effect in ferromagnetic semiconductors

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A magnetization proportional to the square of the electric field is observed in ferromagnetic semiconductors in optical-band fields. It is shown that the effect is proportional to the energy of the s-d exchange interaction of the carriers with the magnetic atoms. The nonlinear magnetization at $\omega \rightarrow q \omega$ is proportional to $q^4$ and $n_1$, at $\omega = 0$ it is proportional to $q^2$ and is independent of $n$ ($q$ and $\omega$ are the wave vector and frequency of the nonlinear magnetization, while $n$ is the carrier density). Action of two laser beams of equal frequency on a crystal produced a specified inhomogeneous static distribution of the magnetization. The nonlinear magnetization at $\omega \rightarrow q \omega$ is proportional to $q^2 \omega$ and is independent of $n$. Action of two laser beams of equal frequency on a crystal produced an inhomogeneous static distribution of the magnetization.

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1. Landau and Lifshitz have indicated in their monograph[1] that, for certain definite magnetic symmetry classes, a magnetoelectric effect can exist wherein a magnetization (polarization) proportional to the electric (magnetic) field can be produced in a crystal. Dzyaloshinskii[22] has shown subsequently that the antiferromagnetic Cr$_2$O$_3$ has a magnetic symmetry that makes this effect possible, as was subsequently observed experimentally[21] in Cr$_2$O$_3$.

However, magnetization proportional to the square of the electric field can exist in all magnets. We have named this the nonlinear magnetoelectric effect. At sufficient field amplitudes, naturally, the nonlinear effect can be appreciable. We shall investigate below the nonlinear magnetoelectric effect in ferromagnetic semiconductors in optical-band electric fields. Many ferroelectric semiconductors have forbidden bands on the order of 1 eV (see, e.g., the reviews[34–36]) and are by the same token transparent enough in the optical band for which high-power lasers are available.

The physical meaning of the considered effect can be explained in the following manner: It is known that a transverse electric field $E$ excites electron-density oscillations in second order in the field. In ferromagnetic semiconductors, the s-d exchange energy $A_S$, where $S$ is the spin of the magnetic atom, exceeds the Fermi energy $E_F$ of the carriers, up to the highest possible values of the concentration $n$, so that the carrier spins have all the same direction. The $\epsilon$ component $s_\epsilon$ of the electron spin density of these polarized carriers is determined by the electron density, so that the resultant $\Delta s_\epsilon$, which is proportional to $E^2$, alters in turn the effective magnetic field $H_{eff}$ that acts on the spin of the magnetic atoms as a result of the s-d exchange interaction and is proportional to $A$. This produces a nonlinear magnetization proportional to the s-d exchange energy and the square of the electric field. By the same token the experimental observation of this effect can be used to investigate exchange interaction (to measure its magnitude and its dependence on the parameters).

2. We shall consider wide-band ferromagnetic semiconductors in which the widths of the conduction band $W$ is large in comparison with $A_S$. The Hamiltonian of the crystal is written in the form

$$H = H_0 + H_s + H_{\text{cont}},$$

(1)

here


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where $a^+_\mathbf{k}$, $a_\mathbf{k}$, $b^+_\mathbf{k}$, $b_\mathbf{k}$ are the electron and magnon creation and annihilation operators (the spin indices have the same direction when considering the spin of the Coulomb interaction of the electrons.)

The Hamiltonian (1) is analogous to the one used by Grinin et al. [17] and obtained from the $s-d$ model Hamiltonian by using a canonical transformation of the Fröhlich type, which eliminates the terms linear in the operators of the deviations of the $d$ spins. In addition, we have taken into account in (1) different electron bands, and also recognize that the $s-d$ exchange integral $A$ is generally speaking different in different bands. [18]

We shall seek the nonlinear magnetization determined by the relation

$$M_{\mathbf{z},\mathbf{r}}(\mathbf{r},t) = \sum_{\mathbf{q}} \langle \hat{\mathbf{a}}^+_{\mathbf{q}} \hat{\mathbf{a}}_{\mathbf{q}} \rangle E_\mathbf{q}(\mathbf{r},t) E_\mathbf{q}(\mathbf{r},t) \exp \{ i (q_z + q_y) r - (q_x + q_z) t \}. \quad (5)$$

where $E_\mathbf{q}(\mathbf{r},t)$ are the Fourier components of the electric field and summation over all repeated indices is implied. The expression for the tensor $\chi_{\mathbf{abc}}$ with allowance for symmetrization takes the form (see, e.g., [19]):

$$\chi_{\mathbf{abc}} \left( \omega_0, \omega_0, \omega_0 \right) = \frac{1}{(2\pi)^3} \int d\mathbf{r}_1 \int d\mathbf{r}_2 \exp \left[ i (\omega_0 + \omega_0) t_1 + i \omega_0 t_2 \right] K_{\mathbf{abc}}^{(1)}(t_1, t_2). \quad (6)$$

Using the procedure of many-time Green's functions (as applied to nonlinear effects [9,10]), we find that $\chi_{\mathbf{abc}}$ is determined by the Fourier component of the retarded Green's function $K_{\mathbf{abc}}^{(1)}$ in accordance with the formula

$$\chi_{\mathbf{abc}} \left( \omega_0, \omega_0, \omega_0 \right) = \frac{1}{(2\pi)^3} \int d\mathbf{r}_1 \int d\mathbf{r}_2 \exp \left[ i (\omega_0 + \omega_0) t_1 + i \omega_0 t_2 \right] K_{\mathbf{abc}}^{(1)}(t_1, t_2). \quad (7)$$

where

$$K_{\mathbf{abc}}^{(1)}(t_1, t_2) = \frac{(2\pi)^3 \mu_B^2 e^2}{V \delta^{3/2} \omega_0^2 \omega_0^2} \sum_{\mathbf{k}_1 \mathbf{k}_2} H_{\mathbf{abc}}^{(1)}(t_1, t_2) p_{\mathbf{a},\mathbf{k}_1}(k) p_{\mathbf{b},\mathbf{k}_2}(k); \quad (8)$$

$$H_{\mathbf{abc}}^{(1)}(t_1, t_2) = 0 \delta(t_1-t_2) \left[ [b^+_\mathbf{k}(t_1) b_\mathbf{k}(t_2), a^+_\mathbf{k}(t_1) a_\mathbf{k}(t_2), a^+_\mathbf{k}(t_1) a_\mathbf{k}(t_2) \right]. \quad (9)$$

In (9), the averaging is with the aid of the density matrix $\rho(-\omega_0)$, $V_0$ is the normalization volume, $\mu_B$ is the Bohr magneton, and

$$\theta(t) = \begin{cases} 1 & t \geq 0 \\ 0 & t < 0 \end{cases} \quad (10)$$

The interaction with the electromagnetic field is given by

$$V_{\mathbf{abc}}^{\mathbf{1}} = -\frac{e}{m c} \sum_{\mathbf{k}} b_{\mathbf{k}+\mathbf{q}} A(\mathbf{q},t) p_{\mathbf{c},\mathbf{k}+\mathbf{q}}(k). \quad (11)$$

where $A(\mathbf{r}, t)$ is the vector potential of the field, and ac-

count is taken of interband transitions, since we are considering an interaction with an optical-band field. It should be noted that we have written here the tensor component that determines the nonlinear magnetization $M_{\mathbf{abc}}^{\mathbf{1}}$ directed along the constant magnetic field. The nonlinear magnetization components $M_{\mathbf{abc}}^{\mathbf{1}}$ are not considered at present; for them to appear, the electric field must cause electron spin flips, which is possible when the spin-orbit interaction takes part.

It is necessary to write a chain of coupled equations of motion for the Fourier components $H_{\mathbf{abc}}(\omega_0, \omega_0)$, using the Hamiltonian (1). Since $AS \gg J$, where $J$ is the energy of the direct exchange between the spins of the magnetic atoms, it is necessary to sum terms that yield energy denominators containing only differences of magnon frequencies. These terms correspond to the diagrams containing resonant cross sections and considered by Grinin and Nagaev, [11,12] who obtained the polarization operator and the "magnetic" response to a longitudinal electric field in a ferromagnetic semiconductor. After rather cumbersome calculations (see the Appendix) we obtain

$$\chi_{\mathbf{abc}} \left( \omega_0, \omega_0, \omega_0 \right) = \frac{\mu_B^2}{V_0} \sum_{\mathbf{k}_1 \mathbf{k}_2} m_{\mathbf{k}_1 \mathbf{k}_2} \left[ \frac{\Delta E_{\mathbf{k}_1}(p)}{\Delta E_{\mathbf{k}_2}(p)} \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_1}(p) \right] \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_2}(p) \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_1}(p) \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_2}(p) \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_1}(p) \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_2}(p) \right]. \quad (12)$$

Here $N_{\mathbf{p}}(p)$ is the carrier distribution function in band $i$, and the populations in formula (11) are both determined from the condition

$$\sum_{\omega_0} \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_1}(p) \right] \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_2}(p) \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_1}(p) \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_2}(p) \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_1}(p) \right] \times \left[ \delta(\omega_0 + \omega_0) - \Delta E_{\mathbf{k}_2}(p) \right]. \quad (13)$$

At concentrations exceeding a certain value $T_1$, and a concentration $n > n_0$, both determined from the condition $\Sigma(\omega_0, \omega_0) = 0$, a divergence is obtained; this agrees with the result of Grinin and Nagaev, [11,12] who found that at $T > T_1$ and $n > n_0$ the homogeneous state of a ferromagnetic semiconductor becomes absolutely unstable. The instability condition [11,12] is determined here for external-field frequencies $\omega = 0$, and in our case the same condition takes place at $\omega_0 + \omega_0 = 0$; from $\Sigma(\omega_0, \omega_0, \omega_0) = 1$ we obtain for field frequencies $\omega_0$ in the optical band a value $T_1$ greatly exceeding $T_1$. We assume henceforth that we are considering a region of temperatures $T < T_1$ and of carrier densities such that the state is stable. Thus, for EuS, according to Grinin and Nagaev, $T_1 = 10^2$ K at $n \approx 10^{20}$ cm$^{-3}$.

The foregoing nonlinearity mechanism does not work for dielectrics, since the contribution from the electrons of the completely filled bands can be easily shown to be determined by summation of an expression of the type $[N_{\mathbf{0}}(p) - N_{\mathbf{0}}(p)]/|F(p)|$ in the entire band ($F$ is a certain
function of the momentum and does not depend on the spin direction. The net result is zero.

We integrate in (11) with respect to the electron quasimomentum, after expanding the population difference in a series and retaining the first term of the expansion. When integrating with respect to angle we choose the polar axis to be the vector $\mathbf{q} = \mathbf{q}_s + \mathbf{q}_r$ and employ the relation

$$\cos(q_s, p) = \cos(q_s, q) + \sin(q_s, q) \cos(q + q_s),$$  
(13)

where $\theta$ and $\varphi$ are the polar and azimuthal angles of the vector $p$, and $\varphi_s$ is the azimuthal angle of the vector $\mathbf{q}_s$. At $q_s - q_r$ we have $\cos(q_s, q) = q / 2 q_s$. Since even in the case of strong doping we have $E_F < AS$ and the radius of the Fermi sphere of the carriers of band $\mathbf{n}_0$ is small ($k_F < k_B T$, where $k_B T$ is the reciprocal-lattice vector), we can write with good approximation $C_{\text{mag}} = C_{\text{mag}}^{(0)}$. As a result, the integration with respect to the electron quasimomentum can be carried out directly if the electron spectrum is given.

We proceed to integrate over the magnon quasimomentum. It should be noted that the electron–magnon interaction leads to a renormalization (see, e.g., [14, 15]) of the magnon frequency. We assume first that the relative carrier density $\nu = n / N$ is such that $A^{(0)} = \nu < JS$, and then the renormalization can be disregarded. Integration over the magnon quasimomentum is over the entire Brillouin zone and can be carried out in two limiting cases—low and high frequencies i.e., $\hbar \omega \ll \omega_s$. In the case of strong doping we have

$$\hbar \omega < \hbar \omega_s < \hbar \omega_s' \gg \hbar \omega_s,$$

where $\omega_s$ is the magnon frequency. We assume that $\hbar \omega_s' \gg \hbar \omega_s$, and in the case of high frequencies formula

$$\omega_s = \left( \frac{m_s (m_s + 1)}{m_s + m_o} \right)^{1/2} (\hbar \omega_s'),$$

we obtain after integrating with respect to angle the function

$$2 - \frac{\hbar \omega_s}{\hbar \omega_s^2} \ln \left( \frac{\omega_s^2}{\hbar \omega_s^2} - JS \right) \frac{\hbar \omega_s^2}{\hbar \omega_s^2 - JS \hbar \omega_s^2}$$

and in the case of high frequencies formula (15) gives the square of the small parameter $JS \hbar \omega_s^2 / \hbar \omega_s^2$, whereas no additional discriminating factor appears if only first-order terms of the expansion of the population difference are taken into account. As a result we obtain

$$x_{\omega, \omega_s} = \frac{\Delta \omega \Delta \omega_s}{\hbar^2 \omega} \left( \frac{\hbar \omega_s^2}{\hbar \omega_s^2 - JS \hbar \omega_s^2} \right) \left( \frac{\hbar \omega_s^2}{\hbar \omega_s^2} - JS \right)^{-1}$$

where $f(\omega_s, q) = 1$ at $\hbar \omega_s < JSq$,  

$$f(\omega_s, q) = \frac{JSq (q_s)^{ij}}{\hbar \omega_s \hbar \omega_s} \left( \frac{b_1 + b_2 \hbar \omega_s}{\hbar \omega_s} \right) \left( \frac{b_1 + b_2 \hbar \omega_s}{\hbar \omega_s} \right),$$

or $\hbar \omega_s > JSq$.  

where $b_1$ and $b_2$ are numerical coefficients of the order of unity, $\omega_s = \omega_s + \omega_s'$, $q = \mathbf{q}_s + \mathbf{q}_r$, $\varepsilon_q = \Delta E_{\mathbf{F}}(\mathbf{q}_s')$, $\Omega_{\mathbf{F}}^2 = \Omega_{\mathbf{F}}^2(\mathbf{q}_s')$, in the estimates $\Omega_{\mathbf{F}}^2 \sim \sigma$, and $m_i$ is the mass of the carrier in the band numbered $i$. We assume that $k_B T \gg \mu M_s$ and have left out terms containing the small parameters

$$AS / W k_B T < 1, b_1^2 / m_a < 1.$$

The function $f(\omega_s, q)$ is determined by the quantity $\omega_s = \hbar \omega_s / W k_B T$, which can be called the parameter of the effective spatial dispersion due to the magnons.

In the optical band we encounter the case $q \ll k_F$, which we shall analyze in detail later on. In the two limiting cases with respect to the parameter $W k_F / \omega_s$, where $\omega_s$ is the Fermi velocity of the carriers, we then have

$$x_{\omega, \omega_s} = \frac{\hbar \omega_s}{\hbar \omega_s^2} \left( \frac{m_i + m_o}{m_i} \right)^{1/2} \left( \frac{m_i + m_o}{m_i} \right)^{1/2} \left[ (\tilde{\varepsilon}(\mathbf{q}_s', \omega_s) - \omega_s) / JSq \right] / JSq,$$

where

$$\tilde{\varepsilon}(\mathbf{q}_s', \omega_s) = \frac{\hbar \omega_s}{\hbar \omega_s^2} \left( \frac{m_i + m_o}{m_i} \right)^{1/2} \left( \frac{m_i + m_o}{m_i} \right)^{1/2} \left[ (\tilde{\varepsilon}(\mathbf{q}_s', \omega_s) - \omega_s) / JSq \right].$$

at $\omega_s \ll W k_F$ and

$$x_{\omega, \omega_s} = \frac{\hbar \omega_s}{\hbar \omega_s^2} \left( \frac{m_i + m_o}{m_i} \right)^{1/2} \left( \frac{m_i + m_o}{m_i} \right)^{1/2} \left[ (\tilde{\varepsilon}(\mathbf{q}_s', \omega_s) - \omega_s) / JSq \right],$$

at $\omega_s \ll W k_F$, where $\omega_s$ is the carrier plasma frequency.

In (19) we use the fact that in the optical band $q$ is much smaller than the reciprocal screening radius of the degenerate carriers $\lambda = (6 \pi n_0^2 / W k_F e^3)^{1/2}$, where $e$ is the static dielectric constant.

Thus, in the high-frequency case $q W k_F / \omega_s \ll 1$ the effect is proportional to $n_i$. In the low-frequency case $q W k_F / \omega_s \gg 1$ it is independent of the carrier density $n_i$, being due to the relation $\varepsilon(\omega_s, q) \sim \hbar q^2 / \omega_s^2 - \hbar q^2 / \omega_s^2$ at $q \ll \Lambda_q$. It can be shown that in the case of nondegenerate carriers at $q W k_F / \omega_s \gg 1$ the effect does not depend on $n$ at $n > n_{\text{crit}}$, where $n_{\text{crit}}$ is determined from the condition $\omega_s^2 / W k_F \ll k_F T$, where $\omega_s$ is the optical Debye screening radius and $W k_F$ is the thermal velocity of the carriers.

We note that the case $\omega_s = 0$ can be realized (for details see below); on the other hand, a nonzero frequency of the nonlinear magnetization is obtained when two lasers with different frequencies $\omega_s$ and $\omega_s'$ are used for the irradiation, so that in the experiment the minimal value $\omega_{\text{mix}} = |\omega_s - \omega_s'| \text{min}$ is determined, obviously, by the condition that $\omega_s / \omega_s' < \omega_{\text{mix}}$.

G. M. Genkin 1976

it is necessary to have \( \omega_1/\omega > 10^{-5} - 10^{-6} \), and then, even for the maximum possible wave vector of the nonlinear magnetization \( q = 2n(\omega)/c \) we have \( n_1 \ll 1 \), and by the same token it follows from (16)-(20) in the case of the experimentally realized nonzero frequencies of the nonlinear magnetization that the result is proportional to \( q^3 \); for zero frequencies \( \omega_1 = 0 \) it is proportional to \( q^2 \). Under conditions of strong doping, when the contribution to the magnon frequency from the electron-magnon interaction predominates over the direct exchange interaction of the magnetic atoms \( A^{(0)}, \mu > JS_e \), we present an approximate formula for the case \( \omega_1 = 0 \):

\[
\chi_m(\omega, -\omega) = \frac{5}{6} \frac{\mu g e^2 (b_T)^2}{(n_0)^2} \frac{\sigma_0}{\sigma_0^*} \frac{\varepsilon_0^*}{\varepsilon_0} \frac{\sigma_1}{\sigma_1^*} [1 - 5 (g/b_T)^2].
\]

We note that now \( \chi \sim n^{3/2} \); it can be shown that \( \chi \sim n^{1/2} \) in the high-frequency case. Thus, in the high-frequency case \( q_2 \omega_2/\omega_1 < 1 \) the optimal value of the concentration is \( n_{\text{opt}} = J S_N / A^{(0)} \), and in the low-frequency case it is the entire range \( n_{\text{opt}} < n < n_{\text{opt}} \).

The obtained relation (19) for \( q^2 \) in the low-frequency case \( \omega_2/\omega_1 \) is the result of the screening of the low-frequency electron-density oscillations produced by the external electric fields. It should be noted that this situation takes place for an isotropic carrier spectrum; for the anisotropic spectrum, the screening can be much less (see the work on Raman scattering of light in semiconductors\[15\] and superconductors\[16\]).

3. We consider the case when two laser beams of frequency \( \omega \) are incident on a crystal, with an angle \( \theta \) between them. The result is, in particular, a static nonlinear magnetization, defined by relation (5) with

\[
\chi_m(q_1, -q_1),
\]

and unevenly distributed over the crystal, with a wave-vector \( q = 2\omega/c \) of \( \omega \sin(\theta/2) \), where \( \omega \) is the refractive index of the light. There is also a nonlinear magnetization at frequency \( 2\omega \), but much weaker,\[10\] as follows from (18)-(20).

The value of the static \( M_s^{(2)}(q) \) is proportional to \( q^2 \sim \sin^2(\theta/2) \). This inhomogeneous magnetization can be revealed, in particular, by its diffraction of a sounding light beam, and, as shown by the estimates presented below for the EuS crystal, lasers of rather modest power are needed to produce at \( \theta = \pi \) a value of \( M_s \) such that the intensity of the diffracted light is of the order of the intensity of the probing radiation. Thus it becomes possible to produce a specified inhomogeneous static distribution of the magnetization \( M_s^{(2)} \), the magnitude and wave vector of which are determined by the value of the given angle \( \theta \) between the laser beams. By the same token, \( q \) varies over a rather wide range \( 0 < q < 2\omega_1/\omega \) at \( 0 < \theta < \pi \). This distinguishes the described effect in principle from the procedure used to excite spin waves under conditions of Suhl instability, where one excites spin waves with fixed wave vectors determined from the condition of their parametric excitation.

We consider the case when the crystal is irradiated by two lasers with different frequencies \( \omega_2 \) and \( \omega_1 \). In this case, in particular, a nonlinear magnetization is produced at the difference frequency \( \omega_1 = |\omega_2 - \omega_1| \) and at \( q = |q_1 - q_1| \). Great interest attaches to the case of sufficiently high frequencies of the order of \( \omega_0 (k_B) \). The frequency \( \omega_0 \) and the wave vector \( q \) of the nonlinear magnetization \( M_s^{(2)} \) are not connected in this case\[17\] by any relation whatever (including the dispersion relation of the spin waves), and by the same token \( \omega_0 \) can also be larger than \( \omega_0 (k_B) \). Such a nonlinear magnetization can be revealed at high frequency by its diffraction of the probing light beam, and also by the effect of generation of an electromagnetic field at this frequency (see Sec. 4).

We note that when the difference frequency \( \omega_1 \) is equal to the plasma frequency \( \omega_p \), it follows from (20) that the effect is resonantly increased by a factor \( \omega_p/\Delta \omega \) times, where \( \Delta \omega \) is the width of the plasma line. In this case the electric fields cause electron density oscillations at the natural frequency \( \omega_1 \). Experimental observation of the nonlinear magnetization in this frequency region as the frequency of one of the lasers is varied can serve as a spectroscopic tool, alongside Raman scattering, for the investigation of plasmas (\( \omega_p, \Delta \omega \)).

Our analysis is limited to electric fields not stronger than the critical field \( E_{cr} \sim M_s^{(2)} \chi^{-1/2} \), where the nonlinear magnetization becomes comparable with the saturation magnetization. In this field region \( E \sim E_{cr} \) the analysis becomes inaccurate; nonetheless, one can state that \( E_{cr} \) is of the order of magnitude of that field at which substantial changes take place in the magnetic subsystem of the ferromagnet as a result of the action of a strong electric field.

From the foregoing results it follows that \( E_{cr} \) is minimal when the laser beams of equal frequency are anti-parallel and the laser frequency is close to the width of the forbidden band. The condition \( \omega_0 = E_{cr} \) can be realized in experiment by using dye lasers\[19\] operating in the entire band \( \lambda = 3400-11750 \) Å and tunable over a wide range (~400 Å). Then we have by way of an estimate

\[
E_{cr} \approx \frac{M_s J S_N}{\mu_0 A^{(0)}(q_1) \varepsilon_0} \left( \frac{J S}{k_B T} \right)^{1/2},
\]

where \( \varepsilon_0 - \mu_0 \omega_0 \sim \mu_0 \nu \) and \( \nu \) is the relaxation frequency.

The nonlinear magnetization leads to a number of physical effects, particularly to a nonlinear interaction of the waves and to diffraction of the light. We proceed to consider these effects.

4. We consider the generation of combination-frequency waves by a nonlinear ferromagnet. A difference frequency \( \omega = |\omega_2 - \omega_1| \) is generated by two pump lasers with frequencies \( \omega_2 \) and \( \omega_1 \). The nonlinear wave equation is of the form

\[
\alpha^2 \frac{\partial E}{\partial t} + \frac{\partial^2 E}{\partial x^2} = -\alpha_0 \frac{\partial^2 E}{\partial x^2} \omega \chi M_s^{(1)}.
\]

If the phase velocities of the pump fields are directed along the \( x \) axis, then there is no mixing effect, since the nonlinear source in the wave equation is equal to zero: \( \text{curl} \chi \chi \chi \chi = 0 \). We assume next that the pump
lasers have a linearly polarized electric field \( E_z \) and their wave vectors lie in the xy plane (TE modes in a transversely magnetized medium); in this case we need the tensor component \( \chi_{zzz} \) of the nonlinear magnetization. From the nonlinear wave equation (23), written out for fields at frequencies \( \omega_1, \omega_2, |\omega_3 - \omega_1| \), it follows\(^{23}\) that there are two regimes: total spatial synchronism (in this case the phase velocity of the nonlinear magnetization is equal to the phase velocity of the field at the difference frequency), and the mismatch regime. In the former case the field amplitude at the difference frequency increases linearly\(^{21}\) with the length of the crystal \( l \), like

\[
E_{z}^{\omega_3 - \omega_1} = 2\pi \chi_{zzz}(q)E_{z}^{\omega_1}E_{z}^{\omega_2}l \tag{24}
\]

and in the latter case the maximum amplitude is

\[
E_{z}^{\omega_3 - \omega_1} = 8\pi \chi_{zzz}(q)E_{z}^{\omega_1}E_{z}^{\omega_2} \tag{25}
\]

The wave vector \( q \) of the nonlinear magnetization is chosen here to satisfy the spatial-synchronism condition. This condition can be satisfied if the wave vector of the field at the difference frequency is larger than the minimum value of the wave vector of the nonlinear magnetization:

\[
|q_0 - q_{pr}| > \left( \frac{\pi}{\eta_{\omega_1} \omega_1} \right) \sin^{-1} \left( \frac{2 \omega_{\omega_1}}{\omega_{\omega_1} \omega_1} \right) \tag{26}
\]

If \( |\omega_3 - \omega_1| < |\omega_2 - \omega_1| \), then the condition (26) can usually be satisfied and the spatial synchronism is ensured by the small angle \( \theta \) between the pumping laser beams

\[
\sin \theta = \left( \frac{\omega_{\omega_1} - \omega_1}{\omega_{\omega_1} \omega_1} \right) \sin \left( \frac{2 \omega_{\omega_1}}{\omega_{\omega_1} \omega_1} \right) \tag{27}
\]

We consider sufficiently high difference frequencies, which, however, are much lower than the pump laser frequencies (but the parameter \( a_1 \) is less than unity in this case). It follows then from (16), (17), (18), and (20) at a fixed difference frequency \( \omega_1 \), when the angle \( \theta \) between the laser beams is varied, the nonlinearity varies like \( q \sim \sin^4(\theta/2) \). We then obtain that only for the difference frequency satisfying the relation

\[
|\omega_3 - \omega_1| > \left( \frac{\pi}{\eta_{\omega_1} \omega_1} \right) \sin^{-1} \left( \frac{2 \omega_{\omega_1}}{\omega_{\omega_1} \omega_1} \right) \tag{28}
\]

does the spatial-synchronism regime give a larger radiative power than the mismatch regime at the maximum wave vector of the nonlinear magnetization \( q = 2\pi \omega_{\omega_1}/c \), where the nonlinearity is maximal. In the opposite case, more is radiated in the mismatch regime. This is due to the strong dependence of the nonlinearity on the value of the wave vector. In this sense, the situation differs radically from nonlinear-polarization radiation in nonlinear optics, where the spatial-synchronism regime is always more effective. For field frequencies \( \omega_{\omega_1} < 10^{10} \text{ sec}^{-1} \) and sample dimensions \( l < 1 \text{ cm} \), for \( |\omega_3 - \omega_2| < 10^{14} \text{ sec}^{-4} \) the mismatch regime provides a stronger radiation. The radiation power flux density \( S \) at the difference frequency is then

\[
S_{z}^{\omega_3 - \omega_1} = (4\pi)^2 \chi_{zzz} S_0 / c \tag{29}
\]

5. The inhomogeneous static magnetization \( M_{L}(q) \), due to the action of two lasers of equal frequency can be registered by the diffraction of a probing light of intensity \( P_0 \). By varying the angle \( \theta \) between the laser beams from 0 to \( \pi \), it is possible to observe Raman–Nath diffraction\(^{21}\) at \( \alpha < 1 \), and then Bragg diffusion at \( \alpha > 1 \), when the parameter \( a^{21} \) is given by

\[
a = q_{pr}^2 \sin \theta \sin \left( \frac{\pi}{2 \omega_{\omega_1}} \right) \tag{30}
\]

where \( q_{pr} \) is the wave vector of the probing light of frequency \( \omega_{pr} \), \( l \) are the linear dimensions of the region where the probing light interacts with the radiation. Since the frequency of the probing light is of the order of the frequency of the lasers that produce nonlinear magnetization, it follows that \( a = q_{pr}^2 \sin^4(\theta/2) \) and that \( \alpha < 1 \) at small \( \theta \). At \( l < 1 \text{ cm} \) and \( \alpha \leq 3 \times 10^{-3} \), Raman–Nath diffusion takes place; for angles \( \theta > 10^4 \) we have Bragg diffraction. The Bragg diffraction condition determines the wave vector \( q \) of the nonlinear magnetization in terms of the diffraction angle \( \varphi \):

\[
\sin \varphi = q_{pr}^2 \tag{31}
\]

A connection exists between the angles \( \varphi \) and \( \theta \):

\[
\sin \varphi = \frac{\sin \theta}{2} \tag{32}
\]

where \( q_{pr} \) is the wave vector of the laser light that produces \( M_{L}(q) \).

The light is diffraction by crystal dielectric-constant inhomogeneities \( \Delta c_{ij} \) due to the inhomogeneous magnetization; in first-order approximation \( \Delta c_{ij}(q) \) is determined by \( M(q) \) with the aid of the linear magneto-optical parameter, which is expressed in terms of the angle \( \Phi \) of the Faraday rotation per unit length. It is easy to find that the relative intensity of the diffraction maximum of first order is

\[
P_{z} = \left( \frac{q_{pr}}{\pi \varphi} \right)^2 \chi_{zzz} \tag{33}
\]

6. We proceed now to estimates for the EuS crystals, where, according to Methfessel and Mattis,\(^{41} \) \( j = 0.2 \text{ K} \), \( S = 1.2, \varepsilon_l = 1.51 \text{ eV} \) and \( a = 6 \text{ Å} \), \( AS = 0.5 \text{ eV} \); according to Axe\(^{42} \), \( \epsilon_0 = 11.1, \epsilon_a = 4.7 \). Then for a neodymium laser with \( h\omega = 1.17 \text{ eV} \) at \( T = 4 \text{ K} \) we obtain\(^{41} \) \( \chi(\omega) = 0, \theta = \varphi \) = \( 3 \times 10^{-7} \text{ cgs esu} \), which corresponds to a critical field \( E_{cr}^{(1)} = M_0^{\frac{1}{2}} \chi^{-\frac{1}{2}} = 8 \times 10^4 \text{ cgs esu at } M_0 = 2 \times 10^4 \text{ Oe} \).

If tunable dye lasers are used at \( h\omega = \omega_{pr} \), we have \( E_{cr}^{(1)} \approx 6 \times 10^3 \text{ cgs esu at } \nu = 10^{12} \text{ sec}^{-1} \); these lasers also
operate in the picosecond-pulse regime, in which the field is $E \approx 10^4$ egs esu. We can thus obtain fields on the order of critical, in which radical changes take place in the magnetic subsystem of the ferromagnetic semiconductor.

We consider now the diffraction of the probing light by the static nonlinear magnetization. If $I$ ~ $10^5$ deg/cm and the crystal dimension is $l = 1$ mm, the intensity of the first diffraction maximum $P$ is of the order of the intensity $P_0$ of the probing signal at $E = 10^8$ $10^{-4}$. A field $E = 8$ egs esu corresponds to a nonlinear-magnetization-producing pump-laser power $S \approx 2 \times 10^8$ W/cm$^2$. A neodymium laser can provide such modest powers with enough to spare even in the free-running mode. When the probing light is diffracted by high-frequency nonlinear magnetization-producing pump-laser power $S \approx 2 \times 10^8$ W/cm$^2$, the intensity of the probing signal at $E = 10^{-4}$ $10^{-4}$ mm, a field $E = 3$ mm, we need a field $E = 1 \times 10^{-4}$ $10^{-4}$ sec$^{-1}$, at $P = P_0$ and $l = 3$ mm, we need a field $E = 2 \times 10^8$ W/cm$^2$, which is also obtained quite easily with lasers.

In conclusion, I am grateful to V. M. Genkin for stimulating discussions.

**APPENDIX**

We write down a chain of equations of motion for the Fourier components $H(\omega_1, \omega_2)$ obtained by differentiating (9) with respect to $\tau$, without allowance for $\mathcal{E}_{\text{Col}}$:

$$\dot{H}_{\omega_1 \omega_2}(\tau, \omega_1, \omega_2) = \sum_{\omega_3} \left[ F_{+ \omega_1 \omega_2}(\omega_1, \omega_2) + F_{- \omega_1 \omega_2}(\omega_1, \omega_2) \right],$$

(A.1)

where

$$F_{+ \omega_1 \omega_2}(\tau, \omega_1, \omega_2) = C_{+ \omega_1 \omega_2}(\tau) \delta(\tau) \left\{ \left[ b^* \right]_{\omega_1 \omega_2} a_{\omega_1 \omega_2} \right\},$$

$$F_{- \omega_1 \omega_2}(\tau, \omega_1, \omega_2) = -C_{- \omega_1 \omega_2}(\tau) \delta(\tau) \left\{ \left[ b^* \right]_{\omega_1 \omega_2} a_{\omega_1 \omega_2} \right\}.$$ (A.2)

(A.3)

For the sake of simplicity, the times $(\tau_1, \tau_2)$ pertain respectively to all operators situated on the left of the given time, up to the nearest comma or commutator sign; we have introduced also the notation $q = \omega_1 + \omega_2$, and for simplicity we introduce

$$a_n = a_{n \omega_1}, \quad e_n(k) = e_{n \omega_1}.$$ (A.4)

In the subsequent chain of equations of motion, we take into account the resonant terms whose energy denominators contain differences of only magnon frequencies and are therefore small in comparison with the denominators containing the electron energies. We have

$$\left[ \omega_1 + \frac{1}{2} \epsilon(k) \sum_{\tau} \omega_1 - \omega_\tau \right] F_{+ \omega_1 \omega_2}(\omega_1, \omega_2)$$

$$= S_{+ \omega_1 \omega_2}(\omega_1, \omega_2) - S_{- \omega_1 \omega_2}(\omega_1, \omega_2) + \sum_{\omega_3} F_{+ \omega_1 \omega_2}(\omega_1, \omega_2).$$ (A.5)

We have written out here the chain of equations for the function $F_{+ \omega_1 \omega_2}(\tau_1, \tau_2)$; we can write an analogous chain for $F_{- \omega_1 \omega_2}(\tau_1, \tau_2)$. In (A.4), $S_{+ \omega_1 \omega_2}(\omega_1, \omega_2)$, $S_{- \omega_1 \omega_2}(\omega_1, \omega_2)$, $F_{+ \omega_1 \omega_2}(\omega_1, \omega_2)$ are the Fourier components of the following functions:

$$S_{+ \omega_1 \omega_2}(\omega_1, \omega_2) = \frac{1}{2\pi} \int_0^{2\pi} G_{\omega_1 \omega_2}(k_1, -k_2) \delta(k_2 - (k_2, k_2)),$$

$$0(\tau_1) \left\{ a_{\omega_1 \omega_2} a_{\omega_1 \omega_2} \right\},$$

$$S_{- \omega_1 \omega_2}(\tau_1) = \frac{1}{2\pi} \int_0^{2\pi} G_{\omega_1 \omega_2}(k_1, q_2) \delta(k_2 - (k_2, k_2))(\tau_1),$$

$$0(\tau_2) \left\{ a_{\omega_1 \omega_2} a_{\omega_1 \omega_2} \right\},$$

$$F_{+ \omega_1 \omega_2}(\tau_1, \tau_2) = \left\{ C_{+ \omega_1 \omega_2}(\tau_1) \delta(\tau_1) \right\},$$

$$0(\tau_2) \left\{ a_{\omega_1 \omega_2} a_{\omega_1 \omega_2} \right\},$$

$$F_{- \omega_1 \omega_2}(\tau_1, \tau_2) = \left\{ C_{- \omega_1 \omega_2}(\tau_1) \delta(\tau_1) \right\}.$$ (A.6)

(A.7)

Here $\delta_{\tau}$ is the Kronecker symbol, and

$$\Delta(k) = \begin{cases} 1, & k_1 = 0, \\ 0, & k_1 = 0 \end{cases}.$$ (A.8)

We have furthermore

$$(\omega_1 + \omega_2 - \omega_1 - \omega_2, \omega_1 + \omega_2) F_{+ \omega_1 \omega_2}(\omega_1, \omega_2) = \sum_{\omega_3} F_{+ \omega_1 \omega_2}(\omega_1, \omega_2),$$

where

$$F_{+ \omega_1 \omega_2}(\tau_1, \tau_2) =$$

$$\left\{ C_{+ \omega_1 \omega_2}(\tau_1) \delta(\tau_1) \right\}.$$ (A.9)

(A.10)

From the system (A.1)–(A.8) and from the continued chain of equations for the function $F_{\omega_1 \omega_2}$ with allowance for the "resonant" terms we find that $H(\omega_1, \omega_2)$ takes the form of a sum of an infinite number of terms, and as a result we obtain for it the expression

$$H_{\omega_1 \omega_2}(\omega_1, \omega_2) =$$

$$\sum_{\omega_1, \omega_2} \left\{ S_{+ \omega_1 \omega_2}(\omega_1, \omega_2) + S_{- \omega_1 \omega_2}(\omega_1, \omega_2) - S_{+ \omega_1 \omega_2}(\omega_1, \omega_2) \right\} -$$

$$\sum_{\omega_1, \omega_2} \left\{ X_1(p) - X_1(p - q) \right\} C_{+ \omega_1 \omega_2}(\tau_1, \tau_2) \delta(\tau_1)$$

$$\left\{ \omega_1 + \omega_2 - \omega_1 - \omega_2 \right\} -$$

$$\omega_1 + \omega_2 - \omega_1 - \omega_2 + \omega_2$$

$$\omega_1 + \omega_2 - \omega_1 - \omega_2 + \omega_2.$$ (A.10)

In (A.10), the functions $S$ differ from $S^*$ in that the operators $b^* b_{\omega_1 \omega_2}$ are replaced by $b_{\omega_1 \omega_2}^* b_{\omega_1 \omega_2}$ and $C_{+ \omega_1 \omega_2}^{(\tau_1, \tau_2)}$ is replaced by $C_{+ \omega_1 \omega_2}^{(\tau_1, \tau_2)}$. The expression in the curly brackets in (A.10), and the analogous expressions for $S$ and $C_{+ \omega_1 \omega_2}$, are governed by the resonant terms. It is necessary next to write down an analogous chain of equations of motion for the Fourier components of the functions $S(\omega)$ with allowance for the resonant terms, and it is found here that in the summation over $\tau$ only the terms with $\tau = -\omega$ differ from zero.

So far we have disregarded the Coulomb interaction of the carriers. It can be taken into account both by the employed method of the equations of motion, and by the diagram method in the RPA approximation. However, it can be much more illustratively taken into account by starting from the physical meaning of the problem. It can be shown that, as already noted in the main text, the considered effect is due to the existence of a change in the concentration $n^{(s)} E S$, and next $n^{(s)}$ gives rise to $M^{(s)}$ as a result of the $s - d$ exchange interaction. In this case $n^{(s)}$ has a frequency $\omega_s + \omega_1$ and a wave vector.
It is then well known that the Coulomb interaction can be taken into account by dividing the density change obtained without allowance for this interaction by the longitudinal dielectric constant at the corresponding frequency and with the corresponding wave vector.

We thus obtain after cumbersome calculations the final result, formula (11) of the main text.

1) In first order if the electric field is longitudinal.
2) The effect is determined by the carrier density, and we consider therefore high densities of the carriers, which are furthermore degenerate.
3) Thus, in particular, in the CdCr2Se4 crystal the exchange interaction of the holes with the spins of the magnetic atoms is much lower than that of the electrons.
4) The “magnetic” response (magnetization) of a degenerate electron gas of ferromagnetic metals to an electrostatic field was investigated by Kim et al. [11]
5) The momentum of the electromagnetic field was neglected in comparison with the electron quasimomentum in the interband transition energies. The momentum operator was replaced in (11) by the operator $e\Omega$ of the interband dipole moment of the electron.
6) Since there is at present no exact information on the spectra of the ferromagnetic superconductors, we assume that the spectrum of the holes and electrons is isotropic and quadratic, and that they are coupled by a direct optical transition at $k=0$.
7) For ferromagnetic semiconductors such as EuO and EuS we have $J \sim 10^{5}$ and $JS_{a}/k \sim 10^{2}$ cm/sec.
8) We have left out here, in particular, terms proportional to the difference between the exchange integrals in different bands.
9) In nonlinear optics and in the case of nonlinear polarization, the analogous effect is called rectification of the optical radiation.
10) The behavior of the magnetization at optical frequencies is being extensively investigated (see, e.g., the review of Krishna and Chetkin [17]).
11) In this sense, the effect considered here differs from two-magnon Raman scattering at $\omega(k_{R})$ in two-sublattice magnets; furthermore, two-magnon effects of exchange origin cannot occur in ferromagnets at all.
12) The pump field amplitudes in this case are such that the length $L = 2\pi\chi(\varepsilon_{p}\varepsilon_{s})^{1/2}$ of the nonlinear interaction is much larger than the sample dimensions.
13) We assume in the estimates that $A^{(1)} = A^{(1)}$ and $m_{1} = m_{0}$.

Translated by J. G. Adashko