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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 $\omega \gg v_F q$ is proportional to q^4 and n; at $\omega = 0$ it is proportional to q^2 and is independent of n (q and ω 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 M_z, the magnitude and wave vector of which was determined by the values of the angle θ; the value of q can vary in a wide range from 0 to $2q_L$ when the angle θ is varied from 0 to π, where q_L is the wave vector of the light and θ is the angle between the directions of the laser beams. The distribution of the magnetization can be determined from the diffraction of a sounding light beam; according to estimates, to produce a magnetization such that the intensity of the first diffraction maximum is of the order of the intensity of the sounding light, the required lasers are of quite low power. The nonlinear magnetization leads to a nonlinear interaction of the optical-band waves; generation at the difference frequency by means of a nonlinear ferromagnet is considered and, in contrast to the known results of nonlinear optics, the spatial-synchronism regime turns out to be less effective at difference frequencies lower than some definite value.

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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. Dzyaloshinskiĭ[2] has shown subsequently that the antiferromagnetic Cr_2O_3 has a magnetic symmetry that makes this effect possible, as was subsequently observed experimentally[3] in Cr_2O_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[4–6]) 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.[1)] In ferromagnetic semiconductors, the s–d exchange energy AS, where S is the spin of the magnetic atom, exceeds the Fermi energy E_F of the carriers,[2)] up to the highest possible values of the concentration n, so that the carrier spins have all the same direction. The z component s_z of the electron spin density of these polarized carriers is determined by the electron density, so that the resultant Δs_z, 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 AS. The Hamiltonian of the crystal is written in the form

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{sd} + \mathcal{H}_{\text{Coul}}; \tag{1}$$

here

$$\mathcal{H}_0 = \sum_{n\mathbf{k}} E_n(\mathbf{k}) a_{n\mathbf{k}}^+ a_{n\mathbf{k}} + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} b_{\mathbf{q}}^+ b_{\mathbf{q}}, \tag{2}$$

$$\mathcal{H}_{sd} = \sum_{\mathbf{nklr}} C_{\mathbf{klr}}^{(n)} a_{n\mathbf{k}}^+ a_{n,\mathbf{k}-\mathbf{r}} b_{\mathbf{l}}^+ b_{\mathbf{l}+\mathbf{r}}, \tag{3}$$

$$C_{\mathbf{klr}}^{(n)} = \frac{A^{(n)}}{2N}\left\{\frac{E_n(\mathbf{k}+\mathbf{l})-E_n(\mathbf{k})}{A^{(n)}S+E_n(\mathbf{k}+\mathbf{l})-E_n(\mathbf{k})} + \frac{E_n(\mathbf{k}+\mathbf{l})-E_n(\mathbf{k}-\mathbf{r})}{A^{(n)}S+E_n(\mathbf{k}+\mathbf{l})-E_n(\mathbf{k}-\mathbf{r})}\right\} \tag{4}$$

where $a_{n\mathbf{k}}^+$, $a_{n\mathbf{k}}$, $b_{\mathbf{q}}^+$, $b_{\mathbf{q}}$ are the electron and magnon creation and annihilation operators (the spin indices have been omitted, since the spins of all carriers have the same direction when $AS > E_F$); $\mathcal{H}_{\text{Coul}}$ is the Hamiltonian of the Coulomb interaction of the electrons.

The Hamiltonian (1) is analogous to the one used by Grigin *et al.*[7] 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.[3)] We shall seek the nonlinear magnetization determined by the relation

$$M_a^{NL}(\mathbf{r},t) = \chi_{abc}\begin{pmatrix}\omega_s, & \omega_l\\ \mathbf{q}_s, & \mathbf{q}_l\end{pmatrix} E_b(\omega_s,\mathbf{q}_s) E_c(\omega_l,\mathbf{q}_l) \exp\{i[(\mathbf{q}_s+\mathbf{q}_l)\mathbf{r}-(\omega_s+\omega_l)t]\}, \tag{5}$$

where $\mathbf{E}(\omega,\mathbf{q})$ are the Fourier components of the electric field and summation over all repeated indices is implied. The expression for the tensor χ_{abc} with allowance for symmetrization takes the form (see, e.g., [8]):

$$\chi_{abc}\begin{pmatrix}\omega_s, & \omega_l\\ \mathbf{q}_s, & \mathbf{q}_l\end{pmatrix} = \frac{1}{2}\left\{\varkappa_{abc}\begin{pmatrix}\omega_s, & \omega_l\\ \mathbf{q}_s, & \mathbf{q}_l\end{pmatrix} + \varkappa_{acb}\begin{pmatrix}\omega_l, & \omega_s\\ \mathbf{q}_l, & \mathbf{q}_s\end{pmatrix}\right\}. \tag{6}$$

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

$$\varkappa_{abc}\begin{pmatrix}\omega_s, & \omega_l\\ \mathbf{q}_s, & \mathbf{q}_l\end{pmatrix} = \frac{1}{(2\pi)^2}\int_{-\infty}^{\infty} d\tau_1 \int_{-\infty}^{\infty} d\tau_2 \exp[i(\omega_s+\omega_l)\tau_1 + i\omega_l\tau_2] K_{abc}^{\mathbf{q}_s\mathbf{q}_l}(\tau_1,\tau_2), \tag{7}$$

where

$$K_{zbc}^{\mathbf{q}_s\mathbf{q}_l}(\tau_1,\tau_2) = \frac{(2\pi)^2\mu_B e^2}{V_0\hbar^2\omega_s\omega_l m^2}\sum_{\substack{n_1n_2n_3n_4\\ \mathbf{kk_1k_2}}} H_{\mathbf{kk_1k_2}}^{n_1n_2n_3}(\tau_1,\tau_2)\, p_{n_1n_2}(\mathbf{k}_1)\, p_{n_3n_4}(\mathbf{k}_2); \tag{8}$$

$$H_{\mathbf{kk_1k_2}}^{n_1n_2n_3n_4}(\tau_1,\tau_2) = \theta(\tau_1)\theta(\tau_2)\langle[[b_{\mathbf{k}}^+(\tau_1) b_{\mathbf{k}+\mathbf{q}_s+\mathbf{q}_l}(\tau_1),$$
$$a_{n_1\mathbf{k}_1}^+(0) a_{n_2,\mathbf{k}_1+\mathbf{q}_s}(0)],\ a_{n_3\mathbf{k}_2}^+(-\tau_2) a_{n_4,\mathbf{k}_2+\mathbf{q}_l}(-\tau_2)]\rangle. \tag{9}$$

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

$$\theta(\tau) = \begin{cases}1 & \tau\geqslant 0\\ 0 & \tau<0\end{cases}.$$

The interaction with the electromagnetic field is given by

$$V^{int} = -\frac{e}{mc}\sum_{n_1n_2\mathbf{k}} a_{n_1\mathbf{k}+\mathbf{q}}^+ a_{n_2\mathbf{k}} \mathbf{A}(\mathbf{q},t)\, \mathbf{p}_{n_1n_2}(\mathbf{k}), \tag{10}$$

where $\mathbf{A}(\mathbf{r},t)$ is the vector potential of the field, and account 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_z^{NL} directed along the constant magnetic field. The nonlinear magnetization components $M_\pm^{NL}$ 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 $H(\omega_1,\omega_2)$, 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 Grigin and Nagaev,[11,12] who obtained the polarization operator and the "magnetic" response[4)] to a longitudinal electric field in a ferromagnetic semiconductor. After rather cumbersome calculations (see the Appendix) we obtain

$$\varkappa_{zbc}\begin{pmatrix}\omega_s, & \omega_l\\ \mathbf{q}_s, & \mathbf{q}_l\end{pmatrix} = \frac{\mu_B}{V_0}\sum_{\mathbf{pk}n_0n_1}\frac{m_{\mathbf{k}+\mathbf{q}_s+\mathbf{q}_l}-m_{\mathbf{k}}}{\omega_s+\omega_l-\omega_{\mathbf{k}+\mathbf{q}_s+\mathbf{q}_l}+\omega_{\mathbf{k}}}$$
$$\times[\hbar(\omega_s+\omega_l)-\Delta E_0]^{-1}[\hbar(\omega_s+\omega_l)-\Delta E_1]^{-1}\frac{e^2\Omega_{n_0n_1}^b(\mathbf{p})\,\Omega_{n_1n_0}^c(\mathbf{p})}{\hbar^2\omega_s\omega_l}$$
$$\times\frac{\Delta E_{10}^2(\mathbf{p})}{[\Delta E_{10}^2(\mathbf{p})-(\hbar\omega_l)^2]}\{C_{\mathbf{p},\mathbf{k}-\mathbf{q},-\mathbf{q}}^{(1)}[(N_0(\mathbf{p})-N_0(\mathbf{p}-\mathbf{q}_l))(\hbar\omega_l-\Delta E_{10}(\mathbf{p}))$$
$$+(N_0(\mathbf{p})-N_0(\mathbf{p}-\mathbf{q}_s))(\hbar\omega_l+\Delta E_{10}(\mathbf{p}))](\hbar(\omega_s+\omega_l)-\Delta E_0)$$
$$-C_{\mathbf{p},\mathbf{k}+\mathbf{q},-\mathbf{q}}^{(0)}(N_0(\mathbf{p})-N_0(\mathbf{p}-\mathbf{q}_s-\mathbf{q}_l))(\hbar\omega_l-\Delta E_{10}(\mathbf{p}))(\hbar(\omega_s+\omega_l)-\Delta E_1)\}$$
$$\times(1-\Sigma(\omega_s+\omega_l,\mathbf{q}_s+\mathbf{q}_l))^{-1}(1-\Sigma(\omega_l,\mathbf{q}_l))^{-1}\varepsilon^{-1}(\omega_s+\omega_l,\mathbf{q}_s+\mathbf{q}_l). \tag{11}$$

Here $N_i(\mathbf{p})$ is the carrier distribution function in band i, and the populations in formula (11) are referred to a single band n_0 in which carriers are present; $m_{\mathbf{k}}$ is the magnon distribution function; $\varepsilon(\omega,\mathbf{q})$ is the longitudinal dielectric constant. We have introduced the notation[5)]:

$$\Delta E_i \equiv E_i(\mathbf{p}-\mathbf{q}_s-\mathbf{q}_l)-E_i(\mathbf{p}),$$
$$\Delta E_{10}(\mathbf{p}) \equiv E_1(\mathbf{p})-E_0(\mathbf{p}),$$
$$\Sigma(\omega,\mathbf{q}) = \sum_{\mathbf{pk}}\frac{C_{\mathbf{p}+\mathbf{k},\mathbf{k},\mathbf{q}}^{(0)}C_{\mathbf{p},\mathbf{k}+\mathbf{q},-\mathbf{q}}^{(0)}[N_0(\mathbf{p})-N_0(\mathbf{p}-\mathbf{q})]}{\hbar\omega+E_0(\mathbf{p})-E_0(\mathbf{p}-\mathbf{q})}\frac{m_{\mathbf{k}}-m_{\mathbf{k}-\mathbf{q}}}{\omega+\omega_{\mathbf{k}}-\omega_{\mathbf{k}-\mathbf{q}}}. \tag{12}$$

At temperatures exceeding a certain value T_1 and a concentration $n > n_0$, both determined from the condition $\Sigma(\omega,\mathbf{q}) = 0$, a divergence is obtained; this agrees with the result of Grigin 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 $\omega_s + \omega_l = 0$; from $\Sigma(\omega_l,\mathbf{q}_l) = 1$ we obtain for field frequencies ω_l in the optical band a value T_2 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 Grigin and Nagaev,[11] $T_1 \approx 10$ °K at $n \sim 10^{20}$ 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 $\{N_{0\uparrow}(\mathbf{p}) - N_{0\downarrow}(\mathbf{p})\}f(\mathbf{p})$ 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 quasimomenta, 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 $\mathbf{q}=\mathbf{q}_s+\mathbf{q}_l$ and employ the relation

$$\cos(\widehat{\mathbf{q}_s, \mathbf{p}})=\cos\theta\cos(\widehat{\mathbf{q}_s, \mathbf{q}})+\sin\theta\sin(\widehat{\mathbf{q}_s, \mathbf{q}})\cos(\varphi+\varphi_1), \tag{13}$$

where θ and φ are the polar and azimuthal angles of the vector $\mathbf{p}$, and φ_1 is the azimuthal angle of the vector $\mathbf{q}_s$. At $q_s \sim q_l$ we have $\cos(\widehat{\mathbf{q}_s, \mathbf{q}}) \approx q/2q_s$. 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 n_0 is small ($k_F \ll k_{Br}$, where k_{Br} is the reciprocal-lattice vector), we can write with good approximation $C_{\beta\mathbf{kq}} \approx C_{0\mathbf{kq}}$. As a result, the integration with respect to the electron quasimomenta can be carried out directly if the electron spectrum is given.[6)]

We proceed to integrate over the magnon quasimomenta. It should be noted that the electron–magnon interaction leads to a renormalization (see, e.g., [14,6]) of the magnon frequency. We assume first that the relative carrier density $\nu \equiv n/N$ 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., $\hbar(\omega_s+\omega_l) \ll JSa|\mathbf{q}_s+\mathbf{q}_l|$ and $\hbar(\omega_s+\omega_l) \gg JSa|\mathbf{q}_s+\mathbf{q}_l|$, respectively, where a is the constant of the lattice, assumed to be primitive cubic. In the case $\hbar(\omega_s+\omega_l) \gg JSa|\mathbf{q}_s+\mathbf{q}_l|$ the difference between the magnon populations must be expanded in a series accurate to second-order terms, for if only first-order terms are taken into account

$$m_{\mathbf{k}+\mathbf{q}}-m_{\mathbf{k}} \approx m_{\mathbf{k}}(m_{\mathbf{k}}+1)\,2JSa^2\mathbf{kq}/k_BT \tag{14}$$

we obtain after integrating with respect to angle the function

$$2-\frac{\hbar(\omega_s+\omega_l)}{JSa^2kq}\ln\left|\frac{\hbar(\omega_s+\omega_l)+JSa^2kq}{\hbar(\omega_s+\omega_l)-JSa^2kq}\right| \tag{15}$$

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

$$\begin{aligned}\varkappa_{zbc}\begin{pmatrix}\omega_s, \omega_l\\ \mathbf{q}_s, \mathbf{q}_l\end{pmatrix} \approx{}& \frac{5\pi\mu_B n\varepsilon_g^{3/2}(k_BT)^{1/2}e^2\Omega_{12}^{b}\Omega_{21}^{c}}{\hbar^2\omega_s\omega_l E_F[\varepsilon_g^2-(\hbar\omega_l)^2](JS)^{3/2}}\\ \times\Big\{&\varepsilon_g A^{(c)}\Big(2-\frac{1}{m_1-m_0}\Big[\ln\Big|\frac{\hbar k_Fq+\hbar q^2+m_0\omega_1}{\hbar k_Fq-\hbar q^2-m_0\omega_1}\Big|\Big(m_0+\frac{\hbar q^2}{\omega_1}\Big)\\ &\times\Big(m_0+m_1+2\frac{\hbar q^2}{\omega_1}\Big)-2\ln\Big|\frac{\hbar k_Fq+\hbar q^2+m_1\omega_1}{\hbar k_Fq-\hbar q^2-m_1\omega_1}\Big|\Big(m_1+\frac{\hbar q^2}{\omega_1}\Big)^2\Big]\frac{\omega_1}{\hbar k_Fq}\Big)\\ &+\hbar\omega_l\Big[2\Big(\frac{m_1}{m_0}A^{(1)}-A^{(0)}\Big)-\frac{1}{m_1-m_0}\Big(\ln\Big|\frac{\hbar k_Fq+\hbar q^2+m_0\omega_1}{\hbar k_Fq-\hbar q^2-m_0\omega_1}\Big|\\ &\times\Big[2m_1A^{(1)}\Big(1+\frac{\hbar q^2}{m_0\omega_1}\Big)-\Big(m_0+m_1+\frac{2\hbar q^2}{\omega_1}\Big)A^{(0)}\Big]m_0\Big(1+\frac{\hbar q^2}{m_0\omega_1}\Big)\\ &-\ln\Big|\frac{\hbar k_Fq+\hbar q^2+m_1\omega_1}{\hbar k_Fq-\hbar q^2-m_1\omega_1}\Big|\Big[A^{(1)\prime}1+\frac{m_1}{m_0}+2\frac{\hbar q^2}{m_0\omega_1}\Big)\\ &-2A^{(0)}\Big(1+\frac{\hbar q^2}{m_1\omega_1}\Big)\Big]m_1^2\Big(1+\frac{\hbar q^2}{m_1\omega_1}\Big)\Big)\frac{\omega_1}{\hbar k_Fq}\Big]\Big\}f(\omega_1, q)\varepsilon^{-1}(\omega_s+\omega_l, \mathbf{q}_s+\mathbf{q}_l),\end{aligned} \tag{16}$$

where $f(\omega_1, q)=1$ at $\hbar\omega_1 \ll JSaq$,

$$f(\omega_1, q)=4\frac{JS(aq)^2}{\hbar\omega_1}\Big(b_1+\frac{b_2}{5}\frac{k_BT}{\hbar\omega_1}\Big), \quad \text{for } \hbar\omega_1 \gg JSaq, \tag{17}$$

b_1 and b_2 are numerical coefficients of the order of unity, $\omega_1 \equiv \omega_s+\omega_l$, $q \equiv |\mathbf{q}_s+\mathbf{q}_l|$, $\varepsilon_g \equiv \Delta E_{12}(0)$, $\Omega_{12}^b \equiv \Omega_{12}^b(0)$, in the estimates $\Omega_{12} \sim a$, and m_i is the mass of the carrier in the band numbered i. We assume that $k_BT \gg \mu M_0$ and have left out terms containing the small parameters

$$AJS/Wk_BT \ll 1, \ \hbar q^2/mAS \ll 1.$$

The function $f(\omega_1, q)$ is determined by the quantity $a_I \equiv JSaq/\hbar\omega_1$, 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 qv_F/ω, where v_F is the Fermi velocity of the carriers, we then have

$$\varkappa_{zbc}\begin{pmatrix}\omega_s, \omega_l\\ \mathbf{q}_s, \mathbf{q}_l\end{pmatrix} \approx \frac{5\pi\mu_B n\varepsilon_g^{3/2}(k_BT)^{1/2}e^2\Omega_{12}^{b}\Omega_{21}^{c}}{\hbar^2\omega_s\omega_l E_F[\varepsilon_g^2-(\hbar\omega_l)^2](JS)^{3/2}}\varphi(\omega_1, q)f(\omega_1, q), \tag{18}$$

where

$$\begin{aligned}\varphi(\omega_1, q)=&\frac{q^2E_F\varepsilon_0}{6\pi ne^2}\Big\{A^{(0)}+2\frac{\hbar\omega_l}{\varepsilon_g}\Big(A^{(1)}\frac{m_1}{m_0}-A^{(0)}\Big)\\ &-3\Big(\frac{q}{k_F}\Big)^2\Big[A^{(0)}+\frac{2\hbar\omega_l}{6\pi ne^2}\Big(A^{(1)}\frac{m_1}{m_0}-A^{(0)}\Big)\Big]-\Big(\frac{\omega_1}{\hbar k_Fq}\Big)^2\\ &\times\Big[A^{(0)}[(m_0+m_1)^2+m_1^2]+\frac{2\hbar\omega_l}{\varepsilon_g}[m_1(m_0A^{(1)}-m_1A^{(0)})\\ &+(m_0+m_1)^2(A^{(1)}m_1/m_0-A^{(0)})]\Big]\Big\}\end{aligned} \tag{19}$$

at $\omega_1 \ll qv_F$ and

$$\begin{aligned}\varphi(\omega_1, q)=&\frac{2}{3}\Big(\frac{\hbar k_Fq}{\omega_1}\Big)^2\frac{1}{m_0m_1}\Big[A^{(0)}\Big(2+\frac{m_1}{m_0}\Big)\\ &+\frac{\hbar\omega_l}{\varepsilon_g}A^{(1)}\Big(1+2\frac{m_1}{m_0}\Big)\Big](1-\omega_{pl}^2/\omega_1^2)^{-1}\end{aligned} \tag{20}$$

at $\omega_1 \gg qv_F$, where ω_{pl} 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 $\lambda=(6\pi ne^2/E_F\varepsilon_0)^{1/2}$, where ε_0 is the static dielectric constant.

Thus, in the high-frequency case $qv_F/\omega_1 \ll 1$ the effect is proportional to n; in the low-frequency case $qv_F/\omega_1 \gg 1$ it is independent of the carrier density n, this being due to the relation $\varepsilon(\omega_1, q) \sim \lambda^2/q^2 \sim n^{1/3}$ at $q^2 \ll \lambda^2$. It can be shown that in the case of nondegenerate carriers at $qv_T/\omega_1 \gg 1$ the effect does not depend on n at $n > n_{(1)}$, where $n_{(1)}$ is determined from the condition $q^2 \ll k_D^2$, (k_D is the reciprocal Debye screening radius and v_T is the thermal velocity of the carriers).

We note that the case $\omega_1 = 0$ 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 ω_s and ω_l are used for the irradiation, so that in the experiment the minimal value $\omega_{1\min} = |\omega_s - \omega_l|_{\min}$ is determined, obviously, by the condition that ω_1/ω at least exceed the relative width of the laser line. Therefore, when using solid-state lasers,

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 $q = 2\omega n(\omega)/c$ we have[7] $a_I \ll 1$, 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^4; 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 $A^{(0)}\nu > JS$, we present an approximate[8] formula for the case $\omega_1 = 0$:

$$\chi_{xbc}(\omega, -\omega) \sim \frac{5}{6}\frac{\mu_B \varepsilon_g^{-3}(k_BT)^{1/2}\Omega_{12}{}^b\Omega_{21}{}^c e^2\varepsilon_0 q^2[1-5(q/k_F)^2]}{(\hbar\omega)^2[\varepsilon_g{}^2-(\hbar\omega)^2]\nu^{1/2}A^{(0)1/2}}. \quad (21)$$

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 $qv_F/\omega_1 \ll 1$ the optimal value of the concentration is $n_{(2)} \sim JSN/A^{(0)}$, and in the low-frequency case it is the entire range $n_{(1)} < n < n_{(2)}$.

The obtained relation (19) for q^2 in the low-frequency case $\omega_1 \ll qv_F$ 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 ω are incident on a crystal, with an angle θ between them. The result is, in particular, a static[9] nonlinear magnetization, defined by relation (5) with

$$\chi_{zab}\begin{pmatrix}\omega, -\omega\\ q_s, -q_l\end{pmatrix},$$

and unevenly distributed over the crystal, with a wave-vector $q = 2(\omega/c)n(\omega)\sin(\theta/2)$, where $n(\omega)$ is the refractive index of the light. There is also a nonlinear magnetization at frequency 2ω, but much weaker,[10] as follows from (16)–(20).

The value of the static $M_z^{NL}(q)$ 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 $\theta \sim \pi$ a value of M_z 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_z^{NL}, the magnitude and wave vector of which are determined by the value of the given angle θ between the laser beams. By the same token, q varies over a rather wide range $0 < q \lesssim 2\omega n(\omega)/c$ at $0 < \theta \lesssim \pi$. 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 ω_s and ω_l. In this case, in particular, a nonlinear magnetization is produced at the difference frequency $\omega_1 = |\omega_s - \omega_l|$ and at $q = |\mathbf{q}_s - \mathbf{q}_l|$. Great interest attaches to the case of sufficiently high frequencies of the order of $\omega(k_{Br})$. The frequency ω_1 and the wave vector $\mathbf{q}$ of the nonlinear magnetization M_z^{NL} are not connected in this case[11] by any relation whatever (including the dispersion relation of the spin waves), and by the same token ω_1 can also be larger than $\omega(k_{Br})$. 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 ω_1 is equal to the plasma frequency ω_{pl}, it follows from (20) that the effect is resonantly increased by a factor $\omega_{pl}/\Delta\omega$ 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 ω_{pl}. 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 (ω_{pl}, $\Delta\omega$).

Our analysis is limited to electric fields not stronger than the critical field $E_{cr} \sim M_0^{1/2}\chi^{-1/2}$, 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 antiparallel and the laser frequency is close to the width of the forbidden band. The condition $\hbar\omega \approx E_{cr}$ can be realized in experiment by using dye lasers[19] operating in the entire band $\lambda = 3400$–$11\,750$ Å and tunable over a wide range (~400 Å). Then we have by way of an estimate

$$E_{cr\,min}^2 \approx \frac{M_0 JS\hbar\nu}{\mu_B A^{(0)}(aq)^2\varepsilon_0}\left(\frac{JS}{k_BT}\right)^{1/2}, \quad (22)$$

where $\varepsilon_g - \hbar\omega \sim \hbar\nu$ and ν 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 $\omega = |\omega_s - \omega_l|$ is generated by two pump lasers with frequencies ω_s and ω_l. The nonlinear wave equation is of the form

$$\nabla^2\mathbf{E} + \frac{1}{c^2}\frac{\partial^2(\varepsilon\mu\mathbf{E})}{\partial t^2} = -4\pi\frac{\partial}{\partial t}\operatorname{rot}\mathbf{M}^{NL}. \quad (23)$$

If the phase velocities of the pump fields are directed along the z axis, then there is no mixing effect, since the nonlinear source in the wave equation is equal to zero: $\operatorname{curl}\mathbf{M}_z(q_z) = 0$. 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 χ_{zzz} of the nonlinear magnetization. From the nonlinear wave equation (23), written out for fields at frequencies ω_s, ω_l, $|\omega_s-\omega_l|$, it follows[20] 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[12] with the length of the crystal l, like

$$E_0^{\omega_s-\omega_l}=2\pi\chi(q_c)E_0^{\omega_s}E_0^{\omega_l}q_c l, \tag{24}$$

and in the latter case the maximum amplitude is

$$E_0^{\omega_s-\omega_l}=8\pi\chi(q)E_0^{\omega_s}E_0^{\omega_l}. \tag{25}$$

The wave vector q_c 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:

$$n(\omega_s-\omega_l)\,|\omega_s-\omega_l|>|n(\omega_s)\omega_s-n(\omega_l)\omega_l|. \tag{26}$$

If $|\omega_s-\omega_l|\ll\omega_s$, $\omega_l\sim\omega$, then the condition (26) can usually be satisfied and the spatial synchronism is ensured by the small angle θ_c between the pumping laser beams

$$\sin\frac{\theta_c}{2}\approx\frac{|\omega_s-\omega_l|n(\omega_s-\omega_l)}{2\omega n(\omega)}. \tag{27}$$

We consider sufficiently high difference frequencies, which, however, are much lower than the pump laser frequencies (but the parameter a_I is less than unity in this case). It follows then from (16), (17), (18), and (20) that at a fixed difference frequency ω_1, when the angle θ between the laser beams is varied, the nonlinearity varies like $q^4\sim\sin^4(\theta/2)$. We then obtain that only for the difference frequency satisfying the relation

$$|\omega_s-\omega_l|>[\omega n(\omega)]^{4/3}\left(\frac{c}{l}\right)^{1/3}\frac{2}{n(\omega_s-\omega_l)}, \tag{28}$$

does the spatial-synchronism regime give a larger radiative power than the mismatch regime at the maximum wave vector of the nonlinear magnetization $q=2\omega n(\omega)/c$, 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 $\omega\sim10^{15}$ sec^{-1} and sample dimensions $l\sim1$ cm, for $|\omega_s-\omega_l|<10^{14}$ sec^{-1} the mismatch regime provides a stronger radiation. The radiation power flux density S at the difference frequency is then

$$S^{\omega_s-\omega_l}=(4\pi)^3\chi^2S^{\omega_s}S^{\omega_l}/c. \tag{29}$$

5. The inhomogeneous static magnetization $M_z^{NL}(q)$, 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 θ between the laser beams from 0 to π, it is possible to observe Raman–Nath diffraction[21] at $\alpha<1$, and then Bragg diffraction at $\alpha>1$, when the parameter α[21] is given by

$$\alpha=q^2l/q_{pr}n(\omega_{pr}), \tag{30}$$

where q_{pr} is the wave vector of the probing light of frequency ω_{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 $\alpha\sim q_{pr}l\sin^2(\theta/2)$ and that $\alpha<1$ at small θ. At $l\sim1$ cm and $\theta\lesssim3\times10^{-3}$, Raman–Nath diffraction takes place; for angles $\theta>10^{-2}$ we have Bragg diffraction. The Bragg diffraction condition determines the wave vector q of the nonlinear magnetization in terms of the diffraction angle φ:

$$\sin\varphi=q/2q_{pr}. \tag{31}$$

A connection exists between the angles φ and θ:

$$\sin\varphi=\frac{q_0}{q_u}\sin\frac{\theta}{2}, \tag{32}$$

where q_0 is the wave vector of the laser light that produces M^{NL}.

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

$$\frac{P}{P_0}\sim\left(\frac{\Phi l}{n(\omega)}\right)^2\left(\frac{\chi E^2}{M_0}\right)^2. \tag{33}$$

6. We proceed now to estimates for the EuS crystals, where, according to Methfessel and Mattis,[4] $J\approx0.2$ K, $S=\frac{7}{2}$, $\varepsilon_g\approx1.51$ eV and $a\approx6$ Å, $AS\sim0.5$ eV; according to Axe[22] $\varepsilon_0=11.1$, $\varepsilon_\infty=4.7$. Then for a neodymium laser with $\hbar\omega=1.17$ eV at $T\approx4$ °K we obtain[13] $\chi(\omega_1=0,\ \theta=\pi)\approx3\times10^{-7}$ cgs esu, which corresponds to a critical field $E_{cr}^{(1)}=M_0^{1/2}\chi^{-1/2}\approx8\times10^4$ cgs esu at $M_0=2\times10^3$ Oe.

We consider now generation of radiation in the submillimeter band by mixing of two antiparallel laser beams with frequencies ω_s and ω_l, such that $\omega_s-\omega_l\sim10^{13}$ sec^{-1}; in this case $\chi\approx5\times10^{-11}$ cgs esu at a carrier density $n\approx3\times10^{18}$ cm^{-3}. If we use picosecond neodymium pump lasers,[23] when the radiation power flux density can be focused to an order of 10^{11} W/cm^2, then we obtain for the radiation flux density of the crystal at 10^{13} sec^{-1} a value of the order of 10 W/cm^2. If $\omega_s-\omega_l\approx\omega_{pl}$, the radiation power is increased by a factor $(\omega_{pl}/\Delta\omega)^2$.

If tunable dye lasers are used at $\hbar\omega\approx\varepsilon_g$, we have $E_{cr\,min}\sim6\times10^3$ cgs esu at $\nu\sim10^{13}$ sec^{-1}; these lasers also

operate in the picosecond-pulse regime, in which the field is $E\sim 10^4$ cgs 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[6] $\Phi\sim 10^5$ deg/cm and the crystal dimension is $l\sim 1$ mm, the intensity of the first diffraction maximum P is of the order of the intensity P_0 of the probing signal at $E^{(1)}\sim E_{cr}^{(1)}\cdot 10^{-4}$. A field $E^{(1)}\sim 8$ cgs esu corresponds to a nonlinear-magnetization-producing pump-laser power $S\sim 2\times 10^4$ 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 of frequency $\omega\sim 10^{13}$ sec^{-1}, at $P\sim P_0$ and $l\sim 3$ mm, we need a field $E^{(2)}\sim 2\times 10^2$ cgs esu (or $S\sim 10^7$ 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 τ_1, without allowance for $\mathscr{H}_{Coul}$:

$$(\omega_1+\omega_{\mathbf{k}}-\omega_{\mathbf{k}+\mathbf{q}})H_{\mathbf{k}\mathbf{k}_1\mathbf{k}_2}^{n_1n_2n_3n_4}(\omega_1,\omega_2)=\sum_{n_0\mathbf{k}_3\mathbf{r}}\{F_{1\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\omega_1,\omega_2)+F_{1\,n_0\mathbf{k}_3\mathbf{r}}^{-}(\omega_1,\omega_2)\},\tag{A.1}$$

where

$$F_{1\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\tau_1,\tau_2)=C_{\mathbf{k}_3,\mathbf{k}-\mathbf{r},\mathbf{r}}^{(0)}\theta(\tau_1)\theta(\tau_2)\langle[[b_{\mathbf{k}}^{+}b_{\mathbf{k}+\mathbf{q}+\mathbf{r}}a_{n_0\mathbf{k}_3}^{+}a_{n_0\mathbf{k}_3-\mathbf{r}}(\tau_1),\ a_{n_1\mathbf{k}_1}^{+}a_{n_2,\mathbf{k}_1+\mathbf{q}_s}(0)],\ a_{n_3\mathbf{k}_2}^{+}a_{n_4,\mathbf{k}_2+\mathbf{q}_l}(-\tau_2)]\rangle;\tag{A.2}$$

$$F_{1\,n_0\mathbf{k}_3\mathbf{r}}^{-}(\tau_1,\tau_2)=-C_{\mathbf{k}_3,\mathbf{k}+\mathbf{q},\mathbf{r}}^{(0)}\theta(\tau_1)\theta(\tau_2)\tag{A.3}$$
$$\times\langle[[b_{\mathbf{k}-\mathbf{r}}^{+}b_{\mathbf{k}+\mathbf{q}}a_{n_0\mathbf{k}_3}^{+}a_{n_0,\mathbf{k}_3-\mathbf{r}}(\tau_1),\ a_{n_1\mathbf{k}_1}^{+}a_{n_2,\mathbf{k}_1+\mathbf{q}_s}(0)],\ a_{n_3\mathbf{k}_2}^{+}a_{n_4\mathbf{k}_2+\mathbf{q}_l}(-\tau_2)]\rangle.$$

For the sake of simplicity, the times $(\tau_1, 0, -\tau_2)$ 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 $\mathbf{q}\equiv\mathbf{q}_s+\mathbf{q}_l$, and for simplicity we introduce

$$a_{n_i\mathbf{k}}\equiv a_{i\mathbf{k}},\qquad \varepsilon_{n_i}(\mathbf{k})\equiv\varepsilon_i(\mathbf{k}).$$

In the subsequent chain of equations of motion, we take into account[11,12] 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

$$\left[\omega_1+\frac{1}{\hbar}(\varepsilon_0(\mathbf{k}_3)-\varepsilon_0(\mathbf{k}_3-\mathbf{r}))+\omega_{\mathbf{k}}-\omega_{\mathbf{k}+\mathbf{q}+\mathbf{r}}\right]F_{1\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\omega_1,\omega_2)$$
$$=S_{1\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\omega_2)-S_{2\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\omega_2)+\sum_{l_1}F_{2\,n_0\mathbf{k}_3\mathbf{r}l_1}^{+}(\omega_1,\omega_2).\tag{A.4}$$

We have written out here the chain of equations for the function $F_1^{+}(\tau_1,\tau_2)$; we can write an analogous chain for $F_1^{-}(\tau_1,\tau_2)$. In (A.4), $S_1^{+}(\omega_2)$, $S_2^{+}(\omega_2)$, $F_2^{+}(\omega_1,\omega_2)$ are the Fourier components of the following functions:

$$S_{1\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\tau_2)=\frac{i}{2\pi}C_{\mathbf{k}_3,\mathbf{k}-\mathbf{r},\mathbf{r}}^{(0)}\delta_{n_0n_4}\Delta(\mathbf{k}_3-\mathbf{r}-\mathbf{k}_1)$$
$$\times\theta(\tau_2)\langle[a_{0\,\mathbf{k}_3}^{+}a_{2,\mathbf{k}_1+\mathbf{q}_s}b_{\mathbf{k}}^{+}b_{\mathbf{k}+\mathbf{q}+\mathbf{r}}(0),\ a_{3\,\mathbf{k}_2}^{+}a_{4,\mathbf{k}_2+\mathbf{q}_l}(-\tau_2)]\rangle;\tag{A.5}$$

$$S_{2\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\tau_2)=\frac{i}{2\pi}C_{\mathbf{k}_3,\mathbf{k}-\mathbf{r},\mathbf{r}}^{(0)}\delta_{n_0n_2}\Delta(\mathbf{k}_1+\mathbf{q}_1-\mathbf{k}_3)$$
$$\times\theta(\tau_2)\langle[a_{1\,\mathbf{k}_1}^{+}a_{0,\mathbf{k}_3-\mathbf{r}}b_{\mathbf{k}}^{+}b_{\mathbf{k}+\mathbf{q}+\mathbf{r}}(0),a_{3\,\mathbf{k}_2}^{+}a_{4,\mathbf{k}_2+\mathbf{q}_l}(-\tau_2)]\rangle;\tag{A.6}$$

$$F_{2\,n_0\mathbf{k}_3\mathbf{r}l_1}^{+}(\tau_1,\tau_2)=C_{\mathbf{k}_3,\mathbf{k}-\mathbf{r},\mathbf{r}}^{(0)}\theta(\tau_1)\theta(\tau_2)$$
$$\times\langle[[\{C_{\mathbf{k}_3-\mathbf{r},l_1,-\mathbf{r}}^{(0)}a_{0\,\mathbf{k}_3}^{+}a_{0\,\mathbf{k}_3}-C_{\mathbf{k}_3-\mathbf{r},l_1,-\mathbf{r}}^{(0)}a_{0,\mathbf{k}_3-\mathbf{r}}^{+}a_{0,\mathbf{k}_3-\mathbf{r}}\}$$
$$\times b_{\mathbf{k}}^{+}b_{\mathbf{k}+\mathbf{q}+\mathbf{r}}b_{l_1-\mathbf{r}}^{+}b_{l_1-\mathbf{r}}(\tau_1),a_{1\,\mathbf{k}_1}^{+}a_{2,\mathbf{k}_1+\mathbf{q}_s}(0)],a_{3\,\mathbf{k}_2}^{+}a_{4,\mathbf{k}_2+\mathbf{q}_l}(-\tau_2)]\rangle.\tag{A.7}$$

Here δ_{ik} is the Kronecker symbol, and

$$\Delta(\mathbf{k})=\begin{cases}1, & \mathbf{k}=0\\ 0, & \mathbf{k}\neq 0.\end{cases}$$

We have furthermore

$$(\omega_1+\omega_{l_1}-\omega_{l_1-\mathbf{r}}+\omega_{\mathbf{k}}-\omega_{\mathbf{k}+\mathbf{q}+\mathbf{r}})F_{2\,n_0\mathbf{k}_3\mathbf{r}l_1}^{+}(\omega_1,\omega_2)=\sum_{n_3\mathbf{k}_4}F_{3\,n_0n_3\mathbf{k}_3\mathbf{k}_4\mathbf{r}l_1}^{+}(\omega_1,\omega_2),\tag{A.8}$$

where

$$F_{3\,n_0n_3\mathbf{k}_3\mathbf{k}_4l_1}^{+}(\tau_1,\tau_2)=C_{\mathbf{k}_3,\mathbf{k}-\mathbf{r},\mathbf{r}}^{(0)}C_{\mathbf{k}_3-\mathbf{r},l_1,-\mathbf{r}}^{(0)}C_{\mathbf{k}_4,l_1-\mathbf{r},\mathbf{r}}^{(ns)}$$
$$\times\theta(\tau_1)\theta(\tau_2)\langle[[b_{\mathbf{k}}^{+}b_{\mathbf{k}+\mathbf{q}+\mathbf{r}}a_{3\,\mathbf{k}_4}^{+}a_{3,\mathbf{k}_4-\mathbf{r}}(b_{l_1}^{+}b_{l_1-\mathbf{r}}-b_{l_1-\mathbf{r}}^{+}b_{l_1-\mathbf{r}})$$
$$\times(a_{0\,\mathbf{k}_3}^{+}a_{0\,\mathbf{k}_3}-a_{0,\mathbf{k}_3-\mathbf{r}}^{+}a_{0,\mathbf{k}_3-\mathbf{r}})(\tau_1),a_{1\,\mathbf{k}_1}^{+}a_{2,\mathbf{k}_1+\mathbf{q}_s}(0)],a_{3\,\mathbf{k}_2}^{+}a_{4,\mathbf{k}_2+\mathbf{q}_l}(-\tau_2)]\rangle.\tag{A.9}$$

From the system (A.1)–(A.8) and from the continued chain of equations for the function F_3 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

$$H_{\mathbf{k}\mathbf{k}_1\mathbf{k}_2}^{n_1n_2n_3n_4}(\omega_1,\omega_2)$$
$$=\sum_{n_0\mathbf{k}_3\mathbf{r}}[S_{1\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\omega_2)+S_{1\,n_0\mathbf{k}_3\mathbf{r}}^{-}(\omega_2)-S_{2\,n_0\mathbf{k}_3\mathbf{r}}^{+}(\omega_2)-S_{2\,n_0\mathbf{k}_3\mathbf{r}}^{-}(\omega_2)]$$
$$\times\frac{(\omega_1+\omega_{\mathbf{k}}-\omega_{\mathbf{k}+\mathbf{q}})^{-1}}{[\omega_1+\hbar^{-1}(\varepsilon_0(\mathbf{k}_3)-\varepsilon_0(\mathbf{k}_3-\mathbf{r}))+\omega_{\mathbf{k}}-\omega_{\mathbf{k}+\mathbf{q}+\mathbf{r}}]}\Bigg\{1$$
$$-\sum_{l\mathbf{p}}\frac{[N_0(\mathbf{p})-N_0(\mathbf{p}-\mathbf{q})]C_{\mathbf{p}-\mathbf{r},\mathbf{k}_3-\mathbf{r},\mathbf{r}}^{(0)}C_{\mathbf{p},\mathbf{k}_3-\mathbf{r},\mathbf{r}}^{(0)}}{[\omega_1+\hbar^{-1}(\varepsilon_0(\mathbf{p})-\varepsilon_0(\mathbf{p}-\mathbf{q}))]}\frac{m_l-m_{l+\mathbf{q}}}{\omega_1+\omega_{l_1}-\omega_{l_1-\mathbf{r}}+\omega_{\mathbf{k}}-\omega_{\mathbf{k}+\mathbf{q}+\mathbf{r}}}\Bigg\}^{-1}.\tag{A.10}$$

In (A.10), the functions S^{-} differ from S^{+} in that the operators $b_{\mathbf{k}}^{+}b_{\mathbf{k}+\mathbf{q}+\mathbf{r}}$ are replaced by $b_{\mathbf{k}-\mathbf{r}}^{+}b_{\mathbf{k}+\mathbf{q}}$ and $C_{\mathbf{k}_3,\mathbf{k}-\mathbf{r},\mathbf{r}}^{(0)}$ are replaced by $C_{\mathbf{k}_3,\mathbf{k}+\mathbf{q},\mathbf{r}}^{(0)}$. The expression in the curly brackets in (A.10), and the analogous expressions $[1-\Sigma(\omega,\mathbf{q})]^{-1}$ in (11) of the main text, 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 $\mathbf{r}$ only the terms with $\mathbf{r}=-\mathbf{q}$ 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 $n^{(2)}\sim E^2$, and next $n^{(2)}$ gives rise to M^{NL} as a result of the s–d exchange interaction. In this case $n^{(2)}$ has a frequency $\omega_s+\omega_l$ and a wave vector

$\mathbf{q}_s + \mathbf{q}_l$. 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 $CdCr_2Se_4$ crystal the exchange interaction of the holes with the spins of the magnetic atoms is much lower than that of the electrons.[5,6]

[4)]The "magnetic" response (magnetization) of a degenerate electron gas of ferromagnetic metals to an electrostatic field was investigated by Kim *et al.*[13]

[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 $\mathbf{k} = 0$.

[7)]For ferromagnetic semiconductors such as EuO and EuS we have $J \sim 1\,^\circ\mathrm{K}$ and $JSa/\hbar \sim 10^3$ 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 Krinchik and Chetkin[17]).

[11)]In this sense, the effect considered here differs from two-magnon Raman scattering at $\omega(k_{Br})$ in two-sublattice magnets; furthermore, two-magnon effects of exchange origin cannot occur in feromagnets at all.[18]

[12)]The pump field amplitudes in this case are such that the length $L = (2\pi\chi q(E_0^{\omega s}E_0^{\omega l})^{1/2})^{-1}$ of the nonlinear interaction is much larger than the sample dimensions.

[13)]We assume in the estimates that $A^{(0)} = A^{(1)}$ and $m_1 = m_0$.

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