SOVIET PHYSICS JETP VOLUME 26, NUMBER 1 JANUARY, 1968

RAMAN SCATTERING OF LIGHT AND THE FARADAY EFFECT IN MAGNETICALLY ORDERED DIELECTRICS

V. S. L'VOV

Semiconductor Institute, Academy of Sciences of the U.S.S.R.

Submitted December 12, 1966

Zh. Eksp. Teor. Fiz. 53, 163–174 (July, 1967)

We construct a phenomenological theory of Raman scattering of light in magnetically ordered dielectrics with two magnetic sublattices, where the scattering is caused both by a direct interaction of the magnetic field of the wave with the spin system and by an indirect interaction of the electric field of the wave with the spins through spin-orbit interaction. These scattering mechanisms are closely connected with the electrical and magnetic gyrotropy of the medium which is responsible for the Faraday effect. We show that taking into account the rotation of the polarization plane of the incident and the scattered radiation leads to a dependence of the differential extinction coefficient on the size and shape of the sample.

RAMAN scattering (RS) is caused by the interaction of the electromagnetic waves with the natural vibrations of the scattering object. In magnetically ordered crystals the degrees of freedom connected with the spin system are important and they lead to the appearance of additional lines in the spectrum of the scattered radiation.[1-3] From a phenomenological point of view these lines of the spectrum are connected with the modulation of the optical characteristics of the medium by spin waves and therefore the frequencies of the incident and scattered light will differ by the magnitudes of the resonance frequencies of the spin system. The modulation of the permittivity and permeability tensors $\hat{\epsilon}$ and $\hat{\mu}$ of the medium is caused by their dependence on the magnetizations ${}^{\alpha}\mathbf{M}$ of the sublattices of the crystal.

In the present paper we restrict our considerations to merely the linear terms in the expansion of $\hat{\epsilon}$ and $\hat{\mu}$ in powers of ${}^{\alpha}\mathbf{M}$. In a transparent crystal only the off-diagonal, purely imaginary components of the tensors $\hat{\epsilon}$ and $\hat{\mu}$ will be proportional to ${}^{\alpha}\mathbf{M}$; these lead respectively to the electrical and magnetic gyrotropy of the medium. The electrical gyrotropy (ϵ_{xy}, ϵ_{xz}, and ϵ_{yz}) arises from the spin-orbit interaction; here the angle of rotation of the polarization plane of the light over a distance equal to the wavelength $2\pi G_e$ turns out to be of the order of the ratio of the spin-orbit interaction ϵ_{so} to the energy splitting of the levels in the crystal field ϵ_{cr} ($G_e \approx \epsilon_{so}/\epsilon_{cr} \approx 3\times10^{-5}$ to 3×10^{-3}). The magnetic gyrotropy of the medium is caused by the direct interaction of the magnetic field of the incident wave with the magnetization of the crystal; here the corresponding angle $G_m \approx 2\pi\gamma M/\omega$, where $\gamma \approx e\hbar/mc$ is the gyromagnetic ratio and ω the frequency of the incident light ($G_m \approx 3\times10^{-5}$).

The mechanism of the Raman scattering of light by a spin system considered in the present paper consists thus in the modulation of the off-diagonal components of the tensors $\hat{\epsilon}$ and $\hat{\mu}$ by the thermal vibrations of the magnetization ${}^{\alpha}\mathbf{M}$ of the sublattices. We elucidate here the close connection between the Raman scattering of light and the "electrical" $\mathbf{G}_e$ and "magnetic" $\mathbf{G}_m$ Faraday effect.[12]

The extinction coefficient[1)] of the "magnetic" Raman scattering (MRS) h_m is determined by the magnitude of the "magnetic" rotation of the polarization plane of the light:

$$h_m \approx \frac{\omega}{c} G_m^2 \left(\frac{a}{\lambda}\right)^3 \operatorname{cth}\frac{\hbar\Omega}{2kT}, \tag{1a}$$

where ω and λ are the frequency and wavelength of the incident light, a is the lattice constant ($a \approx 5\times10^{-8}$ cm) and Ω the frequency of the uniform precession of the magnetization. For the "electrical" Raman scattering (ERS) we have similarly

$$h_e \approx \frac{\omega}{c} G_e^2 \left(\frac{a}{\lambda}\right)^3 \operatorname{cth}\frac{\hbar\Omega}{2kT}. \tag{1b}$$

[1)]The scattering intensity characterizes the differential extinction coefficient dh which is equal to the ratio of the number of photons scattered into the solid angle $d\theta$ per unit time and unit volume of the medium to the photon current density in the incident light: $h = \int (dh/d\theta)\, d\theta$.

We shall give estimates for the case of a ferrodielectric. Bearing in mind that $G \approx 3 \times 10^{-5}$ to 3×10^{-3} and putting $kT/\hbar\Omega \approx 50$ we find in the optical band of frequencies $h \approx 10^{-8}$ to 10^{-12}. This is only a few orders of magnitude less than the Brillouin scattering in crystals and when lasers are present it can certainly be observed.[3]

When one considers the scattering of light one usually does not take the gyrotropy of a crystal into account, assuming it to be small. In our case the very existence of the scattering is connected with the gyrotropy, and neglect of the rotation of the polarization plane of the incident and the scattered light is admissible only in the case of a sufficiently small sample so that the angle of rotation of the polarization plane ψ over its length is much less than π. When the characteristic dimensions of the crystal are such that $\psi \approx \pi$, the angular characteristics $f = \hbar^{-1}\, dh/d\theta$ and the polarization properties of the scattered light depend on its shape and size. From a physical point of view the occurrence of such "shape effects" is connected with the existence of a preferential direction in the crystal: the direction of the magnetization $\boldsymbol{\delta} = \mathbf{M}/M$. The probability for the scattering of light in a well-defined solid angle $d\theta$ by a small region of the crystal depends on the angle between $\boldsymbol{\delta}$ and the direction of the polarization of the light in it. This angle is different in different points of the sample and the angular characteristics $f = h^{-1} dh/d\theta$ when linearly polarized light is scattered will depend on the size and shape of the sample. This dependence can be neglected for small ($\psi \ll \pi$) and large ($\psi \gg \pi$) samples (see Eq. (20d)). In the latter case f does for linearly polarized light not depend on the direction of the polarization and is the same as f for natural light (Eqs. (16b, c)). For natural and circularly polarized light only the polarization properties of the scattered light will depend on the shape of the sample.

We must note the close analogy between magnetic and electric RS. The position and shape of the MRS and ERS spectral lines turn out to be the same. It may happen that in one frequency range $\omega \approx \omega_1$ the Faraday effect is caused by magnetic gyrotropy, and in another range, when $\omega \approx \omega_2$, by the electric one. In that case MRS dominates for $\omega \approx \omega_1$ and ERS dominates for $\omega \approx \omega_2$. Also

$$h(\omega_1)/\omega_1^4 G^2(\omega_1) \sim h(\omega_2)/\omega_2^4 G^2(\omega_2),$$

and in ferrodielectrics these quantities are exactly the same. Moreover, when natural or circularly polarized light is scattered $f(\omega_1) = f(\omega_2)$. When linearly polarized light is scattered some differences in the properties of MRS and ERS will occur.

In the present paper we consider RS in crystals with two magnetic sublattices. This is connected with the fact that the majority of the crystals which are transparent in the visible region have just such a structure (MnF_2, FeF_2, $RbNiF_3$, and so on[4]).

In conclusion we emphasize that we limit ourselves to the linear terms in the expansion of ϵ in terms of the sublattice magnetizations. We thereby disregard RS connected with the participation of two magnons. In accordance with the conservation laws magnons with any momentum up to the limiting momentum can take part in such a process. A microscopic theory is therefore essentially necessary to describe it which takes, for instance, the spin wave dispersion into account. Two-magnon Raman scattering can occur in first order perturbation theory in the parameter $\epsilon_{ex}/\epsilon_{cr}$ where ϵ_{ex} is the energy splitting of the levels due to exchange interaction. This mechanism of two-magnon RS will not contain additional small parameters as compared to the one-magnon RS which is considered in the present paper from a macroscopic point of view. A microscopic analysis of two-magnon scattering will be the subject of a separate communication.

1. THE WAVE EQUATION IN A GYROTROPIC MEDIUM

Landau and Lifshitz[5] have shown that the macroscopic Maxwell equations in a medium for optical frequencies do not have the usual form

$$\operatorname{rot}\mathbf{E} + \frac{1}{c}\frac{\partial \mathbf{B}}{\partial t} = 0, \quad \operatorname{rot}\mathbf{H} - \frac{1}{c}\frac{\partial \mathbf{D}}{\partial t} = 0.$$

We shall therefore start directly from the Lorentz equation for the microscopic electromagnetic field **e**, **h**. After averaging them we get

$$\operatorname{rot}\mathbf{E} + \frac{1}{c}\frac{\partial \mathbf{B}}{\partial t} = 0, \tag{2a}$$

$$\operatorname{rot}\mathbf{B} - \frac{1}{c}\frac{\partial \mathbf{E}}{\partial t} = \frac{4\pi}{c}(\overline{\rho\mathbf{v}} + \mathbf{j}_s). \tag{2b}$$

Here $\mathbf{E} = \bar{\mathbf{e}}$, $\mathbf{B} = \bar{\mathbf{h}}$, ρ is in the case of dielectrics the density of bound charges, and $\mathbf{v}$ their velocity. We have here on the right-hand side of (2b) added the current $\mathbf{j}_S$ caused by the spin magnetization $\mathbf{M}_S$ of the ferrodielectric.[2)] In the case of a small spin-orbit interaction such a splitting-up of the current in an orbital $\overline{\rho\mathbf{v}}$ and a spin $\mathbf{j}_S$ part is unambiguous for frequencies for which the macroscopic Maxwell equations make sense, and then[6]

[2)] For the sake of simplicity we first of all consider a ferrodielectric. The final results will be equally valid also for crystals with two magnetic sublattices.

$$\mathbf{j}_s = c\,\mathrm{rot}\,\mathbf{M}_s. \tag{3}$$

In contrast, the splitting of the current $\overline{\rho\mathbf{v}}$ into a "polarization" and a "magnetic" part loses clearly its meaning for optical frequencies when the "polarization" current exceeds the "magnetic" one.[5] This follows because for such frequencies the concept of orbital magnetization and of the polarization of the substance loses its unambiguous meaning. In accordance with this we shall by definition assume that

$$\partial\mathbf{P}/\partial t = \overline{\rho\mathbf{v}}. \tag{4}$$

Moreover, we define the vector $\mathbf{D}$ as follows:

$$\frac{\partial}{\partial t}\mathbf{D} = \frac{\partial}{\partial t}(\mathbf{E} + 4\pi\mathbf{P}). \tag{4a}$$

Then

$$\mathrm{div}\,\mathbf{D} = 0. \tag{5}$$

We restrict the considerations to such cubic crystals where $M_{orb} \ll M_S$. Neglecting spatial dispersion we write down the material equation which connects the ω-Fourier components of $\mathbf{D}$ and $\mathbf{E}$ in the form

$$D_\alpha(\omega) = \varepsilon_{\alpha\beta}(\omega, \mathbf{M}_{orb}, \mathbf{M}_s)E_\beta(\omega).$$

Taking into account that the spin-orbit interaction is small we expand $\epsilon_{\alpha\beta}$ in a series in $\mathbf{M}_{orb}$, $\mathbf{M}_S$ ($\mathbf{M} = \mathbf{M}_{orb} + \mathbf{M}_S$, $\mathbf{M} \parallel \mathbf{M}_{orb}$, $\mathbf{M}_S$) and restrict ourselves to the linear terms. We use here the general properties of the tensor $\epsilon_{\alpha\beta}$[7]

$$\varepsilon_{\alpha\beta}^*(\omega, \mathbf{M}_{orb}, \mathbf{M}_s) = \varepsilon_{\alpha\beta}(-\omega, \mathbf{M}_{orb}, \mathbf{M}_s),$$
$$\varepsilon_{\alpha\beta}(\omega, \mathbf{M}_{orb}, \mathbf{M}_s) = \varepsilon_{\beta\alpha}(\omega, -\mathbf{M}_{orb}, -\mathbf{M}_s)$$

and we shall assume that the crystal is transparent: $\epsilon^*_{\alpha\beta} = \epsilon_{\beta\alpha}$. Then

$$\mathbf{E}(\omega) = \frac{\mathbf{D}}{\varepsilon(\omega, 0, 0)} - \frac{4\pi i g(\omega)}{\omega\varepsilon(\omega, 0, 0)}[\mathbf{M}\mathbf{D}(\omega)], \tag{6*}$$

where ϵ and g are real even functions of the frequency ω.

When there is an external electromagnetic field $\mathbf{D}^{ex}$, $\mathbf{B}^{ex}$ present it is necessary to add to the expression for $\mathbf{E}(\omega)$ a nonlinear term caused by the thermal vibrations of the moment $\tilde{\mathbf{M}}(t)$. Bearing in mind that the eigenfrequencies of the spin system $\Omega \ll \omega$ we may assume that up to terms of the order of Ω/ω

$$\mathbf{E} = \frac{\mathbf{D}(\omega)}{\varepsilon} - \frac{4\pi i g}{\omega\varepsilon}[\mathbf{M}\mathbf{D}(\omega)] - \frac{4\pi i g}{\omega\varepsilon}[\tilde{\mathbf{M}}(\Omega)\mathbf{D}^{ex}(\omega^{ex})]. \tag{7}$$

Here

$$\tilde{\mathbf{M}}(\Omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\,\tilde{\mathbf{M}}(t)e^{i\Omega t}, \quad \omega = \omega^{ex} + \Omega.$$

*$[\mathbf{M}\mathbf{D}] \equiv \mathbf{M}\times\mathbf{D}$.

We must add to the Maxwell equations (2) the equation of motion of the magnetic moment. Bearing in mind that $\Omega \ll \omega$ and $\mathbf{M} \approx \mathbf{M}_S$ we have for vibrations of frequency ω

$$\frac{\partial\mathbf{M}_s}{\partial t} = \gamma[\mathbf{M}\mathbf{H}(\omega)] + \gamma[\tilde{\mathbf{M}}(\Omega)\mathbf{H}^{ex}(\omega^{ex})], \tag{8}$$

where γ is the gyromagnetic ratio. From (2)–(8) we get the Maxwell equations for frequency ω. Eliminating $\mathbf{H}$ from them we get a wave equation for $\mathbf{D}$:

$$\nabla^2\mathbf{D} + k_0^2\mathbf{D} + 2i(\mathbf{G}\nabla)\,\mathrm{rot}\,\mathbf{D} = 4\pi\{\mathrm{rot}\,\mathrm{rot}\,[\mathbf{A}\mathbf{D}^{ex}] + \mathrm{rot}\,[\mathbf{C}\,\mathrm{rot}\,\mathbf{D}^{ex}]\},$$
$$k_0^2 = \varepsilon(\omega/c)^2, \tag{9}$$

where the gyration vector $\mathbf{G} = -2\pi\omega^{-1}(g+\gamma)\mathbf{M}$ and the source vectors have the form

$$\mathbf{A}(\Omega) = \frac{g}{\omega}\tilde{\mathbf{M}}(\Omega), \quad \mathbf{C}(\Omega) = \frac{\gamma}{\omega}\tilde{\mathbf{M}}(\Omega).$$

In a dielectric with two sublattices $^1\mathbf{M}$ and $^2\mathbf{M}$ the final Eq. (9) has the same form. Here

$$\omega\mathbf{G} = -2\pi\{(g+\gamma^+)\mathbf{M} + (q+\gamma^-)\mathbf{L}; \tag{10a}$$
$$\omega\mathbf{A}(\Omega) = g\tilde{\mathbf{M}}(\Omega) + q\tilde{\mathbf{L}}(\Omega),$$
$$\omega\mathbf{G}(\Omega) = \gamma^+\tilde{\mathbf{M}}(\Omega) + \gamma^-\tilde{\mathbf{L}}(\Omega), \tag{10b}$$

where

$$2\gamma^\pm = \gamma_1 \pm \gamma_2, \quad \mathbf{M} = {}^1\mathbf{M} + {}^2\mathbf{M}, \quad \mathbf{L} = {}^1\mathbf{M} - {}^2\mathbf{M}$$

and so on, and the phenomenological quantities g and q are real even functions of the frequency ω.

2. THE FARADAY EFFECT

When there is no external electromagnetic field Eq. (9) becomes homogeneous and describes the propagation of plane waves $\mathbf{D} \sim e^{i\mathbf{k}\cdot\mathbf{r}}$ in the medium. It has a non-trivial solution for two values of $\mathbf{k}$:

$$k^\pm = k_0[1 \mp 2\boldsymbol{\alpha}\mathbf{G}]^{-1/2} = k_0[1 \pm \boldsymbol{\alpha}\mathbf{G}], \quad \boldsymbol{\alpha} = \mathbf{k}/k,$$

corresponding to the propagation of waves which are right-hand and left-hand circularly polarized. The rotation of the plane of polarization per unit length of the sample is $k_0\boldsymbol{\alpha}\cdot\mathbf{G}$ and the dimensionless parameter $2\pi\mathbf{G} \ll 1$ is the angle of rotation of the polarization plane for $\mathbf{G} \parallel \boldsymbol{\alpha}$ over a distance equal to the wavelength.

In antiferromagnetics which are odd in Turov's terminology[8] we have $\mathbf{L} \to -\mathbf{L}$ under an inversion $\mathbf{M} \to \mathbf{M}$. Then g is a tensor and q a pseudotensor of second rank ($q = 0$). Moreover, $\gamma_1 = \gamma_2$, i.e., $\gamma^- = 0$ and $\mathbf{M} = 0$ without an external magnetic field. Hence in pure antiferromagnetics $\mathbf{G} = 0$

(see (10a)) and there is no Faraday effect without an external magnetic field. In even antiferromagnetics under an inversion $\mathbf{M} \to \mathbf{M}$ and $\mathbf{L} \to \mathbf{L}$ and weak ferromagnetism, $q \neq 0$, is possible and there is a Faraday effect caused both by ferromagnetic and by antiferromagnetic moments.

3. THE GREEN FUNCTION OF THE WAVE EQUATION IN A GYROTROPIC MEDIUM

We shall solve the inhomogeneous Eq. (9) by a Green function method. To do this we consider first the corresponding equation with point sources:

$$\nabla^2\mathbf{D} + k_0^2\mathbf{D} + 2i(\mathbf{G}\nabla)\,\mathrm{rot}\,\mathbf{D} = -4\pi\mathbf{n}\delta(\mathbf{r}-\mathbf{r}'), \quad |\mathbf{n}| = 1,$$

in the k-representation it becomes algebraic and can easily be solved. Taking the inverse Fourier transformation[3)] we get the Green function of the wave Eq. (8) in a gyrotropic medium:

$$\begin{aligned}\mathbf{D}(\mathbf{r}-\mathbf{r}') = \frac{1}{2|\mathbf{r}-\mathbf{r}'|}\{&(\mathbf{n} - i[\boldsymbol{\alpha}\mathbf{n}] - \boldsymbol{\alpha}(\boldsymbol{\alpha}\mathbf{n}))\exp ik_\alpha^+|\mathbf{r}-\mathbf{r}'| \\ &+ (\mathbf{n} + i[\boldsymbol{\alpha}\mathbf{n}] - \boldsymbol{\alpha}(\boldsymbol{\alpha}\mathbf{n}))\exp ik_\alpha^-|\mathbf{r}-\mathbf{r}'| \\ &+ 2\boldsymbol{\alpha}(\boldsymbol{\alpha}\mathbf{n})\exp ik_0|\mathbf{r}-\mathbf{r}'|\},\end{aligned} \tag{11}$$

where $k_\alpha^\pm = k_0(1 \pm \boldsymbol{\alpha}\cdot\mathbf{G})$ and $\boldsymbol{\alpha} = (\mathbf{r}-\mathbf{r}')/|\mathbf{r}-\mathbf{r}'|$ is the direction of propagation of the scattered light. In a non-gyrotropic medium, when $G = 0$, the quantity $k^\pm = k_0$ and Eq. (11) goes over into the well-known Green function of the d'Alembert equation:

$$D(\mathbf{r}-\mathbf{r}') = \frac{\exp\{ik|\mathbf{r}-\mathbf{r}'|\}}{|\mathbf{r}-\mathbf{r}'|}.$$

4. LIGHT SCATTERING IN A GYROTROPIC MEDIUM

Using the Green function (11) we can easily write down the solution of Eq. (9) in the wave zone ($\mathbf{r} \gg \mathbf{r}'$; $\mathbf{r}'$ varies within the limits of the source). Integrating the expression obtained by parts we get after simple but tedious calculations

$$D(\mathbf{r}) = \frac{\varepsilon\omega^2}{2c^2r}[\boldsymbol{\alpha}[\boldsymbol{\alpha}\mathbf{K}(\mathbf{r})]], \tag{12a}$$

where

$$\begin{aligned}\mathbf{K}(\mathbf{r}) = {}&\exp(i\mathbf{k}_\alpha^+\mathbf{r})\int d^3r'(\mathbf{P}^+ - i[\boldsymbol{\alpha}\mathbf{P}^+])\exp\{-ik_\alpha^+(\boldsymbol{\alpha}\mathbf{r}')\} \\ &+ \exp(i\mathbf{k}_\alpha^-\mathbf{r})\int d^3r'(\mathbf{P}^- + i[\boldsymbol{\alpha}\mathbf{P}^-])\exp\{-ik_\alpha^-(\boldsymbol{\alpha}\mathbf{r}')\},\end{aligned} \tag{12b}$$

$$\mathbf{P}^\pm = [\mathbf{C}\mathbf{D}^{ex}] \pm i[\mathbf{A}[\boldsymbol{\beta}\mathbf{D}^{ex}]], \qquad \boldsymbol{\beta} = \mathbf{k}^{ex}/k^{ex}. \tag{12c}$$

[3)] It is convenient to write the integral over d^3k in a spherical coordinate system, choosing the z-axis along the direction of $\mathbf{r}-\mathbf{r}'$. Repeatedly integrating it over $x = \cos\theta$ we get an expansion of $\mathbf{D}\cdot(\mathbf{r}-\mathbf{r}')$ in inverse powers of $k_0|\mathbf{r}-\mathbf{r}'| \gg 1$ and we restrict ourselves to the first term. After that the integration over φ and $|\mathbf{k}|$ is elementary.

Characteristic for the scattered radiation is the Hermitean second rank tensor

$$I_{ij}(\omega) = \varepsilon^{-2}\langle D_iD_j\rangle_\omega,$$

where

$$\langle D_iD_j\rangle\,\delta(\omega+\omega') = \overline{D_i(\omega)D_j(\omega')},$$

while the bar indicates averaging over the time. In these equations i and j take on the values x and y and we assume that the scattered light is propagated along the z-axis. Using (12a) one shows easily that

$$I_{ij}(\omega) = \frac{\omega^4}{4c^4r^2}\langle K_iK_j\rangle_\omega. \tag{13}$$

The expressions for K_x and K_y which are of interest to us can be obtained from (12b):

$$\begin{aligned}K_x &= \mathscr{K}^+\exp(i\mathbf{k}_\alpha^+\mathbf{r}) + \mathscr{K}^-\exp(i\mathbf{k}_\alpha^-\mathbf{r}), \\ -iK_y &= \mathscr{K}^+\exp(i\mathbf{k}_\alpha^+\mathbf{r}) - \mathscr{K}^-\exp(i\mathbf{k}_\alpha^-\mathbf{r}).\end{aligned} \tag{14}$$

$\mathscr{K}^\pm$ has a very simple form if the incident light is circularly polarized:

$$(D^{ex})^\pm = D_x^{ex} \pm iD_y^{ex} = D^\pm\exp\{ik_\beta^\pm(\boldsymbol{\beta}\mathbf{r})\}.$$

In contrast to (13) and (14) here the z-axis is $\parallel\boldsymbol{\beta}$, the x-axis $\parallel[\boldsymbol{\alpha}\times\boldsymbol{\beta}]$, and $k_{\beta^\pm} = k_0(1 + \boldsymbol{\beta}\cdot\mathbf{G})$. Then

$$2\mathscr{K}^+(D^\pm) = D^\pm\int d^3rS^\pm\exp\{ik_\beta^\pm(\boldsymbol{\beta}\mathbf{r}) - ik_\alpha^+(\boldsymbol{\alpha}\mathbf{r})\},$$

$$\begin{aligned}2\mathscr{K}^-(D^\pm) &= D^\pm\int d^3rT^\pm\exp\{ik_\beta^\pm(\boldsymbol{\beta}\mathbf{r}) - ik_\alpha^-(\boldsymbol{\alpha}\mathbf{r})\}, \\ -S^\pm &= (\mathbf{A} \pm \mathbf{C},\ [\boldsymbol{\alpha}\boldsymbol{\beta}] + i(\boldsymbol{\alpha} \pm \boldsymbol{\beta})), \\ -T^\pm &= (\mathbf{A} \mp \mathbf{C},\ [\boldsymbol{\alpha}\boldsymbol{\beta}] - i(\boldsymbol{\alpha} \mp \boldsymbol{\beta})).\end{aligned} \tag{15}$$

From this we easily obtain expressions for the correlators we need. E.g.,

$$4\langle\mathscr{K}^+(D^\pm)\mathscr{K}^+(D^\pm)\rangle_\omega = V(D^\pm)^2\langle S^\pm S^\pm\rangle_{\omega^{ex}-\omega,\,\mathbf{q}},$$

$$4\langle\mathscr{K}^+(D^\pm)\mathscr{K}^-(D^\pm)\rangle_\omega = (D^\pm)^2\langle T^\pm S^\pm\rangle_{\omega^{ex}-\omega,\,\mathbf{q}} \times \int_V d^3r\exp\{2ik_0(\boldsymbol{\alpha}\mathbf{G})(\boldsymbol{\alpha}\mathbf{r})\}.$$

Here V is the volume of the scattering medium, the integral is taken over that volume, $\mathbf{q} = k_0(\boldsymbol{\alpha} - \boldsymbol{\beta})$, and

$$\langle S^\pm S^\pm\rangle_{\Omega,\,\mathbf{q}} = \int d^3r\,\langle S^\pm S^\pm\rangle_\Omega\exp(i\mathbf{q}\mathbf{r}).$$

For the sake of simplicity we shall assume in the following that one kind of scattering, e.g., magnetic scattering ($\mathbf{A} \neq 0$, $\mathbf{C} = 0$) dominates.

The differential extinction coefficient $dh/d\theta$ is by definition equal to

$$\frac{dh(\omega)}{d\theta} = \frac{\varepsilon^2R^2\,\mathrm{Sp}\,I(\omega)}{V|\mathbf{D}^{ex}|^2}.$$

If the incident light is circularly polarized ($D^{ex\pm} \neq 0$), then

$$\frac{dh^{\pm}(\omega)}{d\theta}=\frac{\varepsilon^2\omega^4}{4c^4}\left\{\sum_{j=1}^{3}\langle A_jA_j\rangle_{\Omega,\,\mathbf{q}}\pm 2\,\mathrm{Im}\,\langle A_3A_1\rangle_{\Omega,\,\mathbf{q}}\right\}. \quad (16a)$$

Here $\omega = \omega^{ex} + \Omega$ and

$$A_1=\mathbf{A}\,[\boldsymbol{\alpha\beta}],\quad A_2=\mathbf{A}\boldsymbol{\beta},\quad A_3=\mathbf{A}\boldsymbol{\alpha}.$$

In natural light right-hand and left-hand polarization occur with equal probability. Therefore

$$\frac{dh(\omega)}{d\theta}=\frac{1}{2}\left(\frac{dh^+}{d\theta}+\frac{dh^-}{d\theta}\right)=\frac{\varepsilon^2\omega^2}{4c^4}\sum_{j=1}^{3}\langle A_jA_j\rangle_{\Omega,\,\mathbf{q}}. \quad (16b)$$

Light which is linearly polarized with an angle φ to the direction of $[\alpha\beta]$ at the origin is a superposition of left-hand and right-hand polarized waves, and $D^{\pm} = D\,e^{\pm i\varphi}$. Using (15) and (16a) we get for linearly polarized light

$$\frac{dh_{\varphi}(\omega)}{d\theta}=\frac{\varepsilon^2\omega^4}{4c^4}\left\{\sum_{j=1}^{3}\langle A_jA_j\rangle_{\Omega,\,\mathbf{q}}+\frac{2}{V}[\langle A_1A_1\rangle_{\Omega,\,\mathbf{q}} - \langle A_2A_2\rangle_{\Omega,\,\mathbf{q}}+\langle A_3A_3\rangle_{\Omega,\,\mathbf{q}}]\int_V d^3r\cos 2(k_0(\mathbf{G}\boldsymbol{\beta})(\boldsymbol{\beta}\mathbf{r})+\varphi) - \frac{2}{V}[\langle A_1A_2\rangle_{\Omega,\mathbf{q}}+\langle A_2A_1\rangle_{\Omega,\mathbf{q}}]\int_V d^3r\sin 2(k_0(\mathbf{G}\boldsymbol{\beta})(\boldsymbol{\beta}\mathbf{r})+\varphi)\right\}. \quad (16c)$$

The extinction coefficient for the scattering of linearly polarized light depends thus on the size and shape of the sample. When the polarization plane of the incident light undergoes many rotations along the length of the sample, one can neglect the last terms in (16c) and the extinction coefficients of linearly polarized (16c) and natural light (16b) are the same. By averaging (16c) over the angle φ we obtain again (16b).

We obtain similar results in the case when not the magnetic but the electrical Raman scattering ($\mathbf{A} = 0$, $\mathbf{C} \neq 0$) dominates: to obtain $dh^{\pm}/d\theta$ and $dh^{\varphi}/d\theta$ we must in Eqs. (16) replace $\mathbf{A}$ by $\mathbf{C}$ and change the sign to its opposite in front of the integrals in (16c).

The extinction coefficients of MRS and ERS can in magnetically ordered dielectrics thus be expressed in terms of the correlators of the ferromagnetic and antiferromagnetic moments:

$$\langle \tilde{M}_i\tilde{M}_k\rangle_{\Omega,\,\mathbf{q}},\quad \langle \tilde{M}_i\tilde{L}_k\rangle_{\Omega,\,\mathbf{q}},\quad \langle \tilde{L}_i\tilde{L}_k\rangle_{\Omega,\,\mathbf{q}}.$$

To evaluate them it is necessary to make well-defined assumptions about the kind of the magnetic structure of the ferrite.

5. CALCULATION OF THE CORRELATORS

Using the theory of non-thermodynamic fluctuations of several quantities[9] one can show that

$$\langle {}^{\alpha}\tilde{M}_i{}^{\beta}\tilde{M}_k\rangle_{\Omega,\mathbf{q}}=\frac{i\hbar}{4\pi}(\chi_{ki}^{*\beta\alpha}(\Omega,\mathbf{q})-\chi_{ik}^{\alpha\beta}(\Omega,\mathbf{q}))\,\mathrm{cth}\frac{\hbar\Omega}{2T}. \quad (17)$$

Here $\chi_{ik}^{\alpha\beta}(\Omega, \mathbf{q})$ is the magnetic susceptibility tensor of the α-sublattice in the case where the magnetic field with frequency Ω and wavevector $\mathbf{q}$ acts only upon the β-sublattice. When there is no interaction between the sublattices, $\chi_{ik}^{\alpha\beta} \approx \delta_{\alpha\beta}$ and we are led to an obvious result: there is no correlation between the sublattices.

We shall evaluate $(\chi_{ik}^{\alpha\beta})'' = (1/2i)(\chi_{ik}^{\alpha\beta} - \chi_{ki}^{*\beta\alpha})$ in the case when there are no losses. We shall assume that ${}^{\alpha}\mathbf{M} \to {}^{\alpha}\mathbf{M} + {}^{\alpha}\mathbf{m}$, ${}^{\alpha}\mathbf{m} \sim e^{i(\mathbf{q}\cdot\mathbf{r} - \Omega t)}$, the exchange fields are $\lambda^2\mathbf{M}$, $-\lambda^2\mathbf{M}$, and the internal anisotropy fields ${}^1\mathbf{H}_A$, ${}^2\mathbf{H}_A$ are directed along the axis of symmetry of the crystal and act in opposite directions and that the external field $\mathbf{H}_0$ is directed along the same axis (z-axis). For the sake of simplicity we shall not take into account the static demagnetizing field. However, we shall take into account the demagnetizing field of the spin wave $\mathbf{H}_p$ which is important for evaluating the correlators for frequencies comparable to $4\pi\gamma M$.[10]

The equations of motion have the form

$$-i\Omega{}^{\alpha}\mathbf{m}=\gamma_{\alpha}[({}^{\alpha}\mathbf{M}+{}^{\alpha}\mathbf{m}),\quad ({}^{\alpha}\mathbf{H}+{}^{\alpha}\mathbf{h})]$$

where

$${}^1\mathbf{H}=\mathbf{H}_0+\mathbf{H}_p+{}^1\mathbf{H}_A+\lambda({}^2\mathbf{M}+{}^2\mathbf{m}),$$

$${}^2\mathbf{H}=\mathbf{H}_0+\mathbf{H}_p+{}^2\mathbf{H}_A+\lambda({}^1\mathbf{M}+{}^1\mathbf{m}),$$

$$\mathbf{H}_p=-4\pi\frac{\mathbf{q}(\mathbf{q}{}^1\mathbf{m}+\mathbf{q}{}^2\mathbf{m})}{q^2}.$$

Here ${}^{\alpha}\mathbf{h} \sim e^{i(\mathbf{q}\cdot\mathbf{r}-\Omega t)}$ is a high-frequency field acting upon the α-sublattice. In the usual consideration of ferromagnetic resonance[11] there is no necessity to separate ${}^1\mathbf{h}$ and ${}^2\mathbf{h}$ as we are interested in the quantity $\chi_{ik}(\Omega)$ determined by the equation

$$({}^1m_i+{}^2m_i)=\chi_{ik}({}^1h_k+{}^2h_k).$$

However, we are interested in the susceptibility $\chi_{ik}^{\alpha\beta}$: $m_i^{\alpha} = \chi_{ik}^{\alpha\beta}\,{}^{\beta}h_k$ (there is here no summation over k or β). In the initial equations of motion we have not taken the energy losses into account and, of course, when solving them we get the Hermitean part of the tensor $\chi_{ik}^{\alpha\beta}$. We find its anti-Hermitean part from dispersion relations.[9] Substituting this expression into (17) we get the correlators $\langle {}^{\alpha}M_i\,{}^{\beta}M_k\rangle$ through which in an obvious way we can express the correlators $\langle\tilde{M}_i\,\tilde{M}_k\rangle$, $\langle\tilde{M}_i\,\tilde{L}_k\rangle$, and $\langle\tilde{L}_i\,\tilde{L}_k\rangle$.

It is convenient to write down separately the final expressions for them for a ferromagnetic when we can neglect the external field $\mathbf{H}_0$ and the anisotropy field $\mathbf{H}_A$ compared with the difference between the exchange fields for the sublattices and for an antiferromagnetic when it is impossible to do so.

The correlators for a ferromagnetic far from the compensation points have the form:[4)]

$$\langle \tilde{M}_i \tilde{M}_k \rangle_{\Omega\ q\to 0} = \frac{\pi\hbar\gamma^+}{M} \operatorname{cth} \frac{\hbar\Omega}{2T} \{M^2 \Omega_{ik}{}^f \delta(\Omega^2 - \Omega_f{}^2) + {}^1M^2 M(\gamma^-/\gamma^+)^2 \Omega_{ik}{}^e \delta(\Omega^2 - \Omega_e{}^2)\},$$

$$\langle \tilde{M}_i \tilde{L}_k \rangle_{\Omega,\ q\to 0} = \langle \tilde{L}_i \tilde{M}_k \rangle_{\Omega,\ q\to 0} = \frac{\pi\gamma^+\hbar}{M} \operatorname{cth} \frac{\hbar\Omega}{2T} \{ML \Omega_{ik}{}^f \delta(\Omega^2 - \Omega_f{}^2) + {}^1M^2 M(\gamma^-/\gamma^+) \Omega_{ik}{}^e \delta(\Omega^2 - \Omega_e{}^2)\},$$

$$\langle \tilde{L}_i \tilde{L}_k \rangle_{\Omega,\ q\to 0} = \frac{\pi\hbar\gamma^+}{M} \operatorname{cth} \frac{\hbar\Omega}{2T} \{L^2 \Omega_{ik}{}^f \delta(\Omega^2 - \Omega_f{}^2) + {}^1M^2 M \Omega_{ik}{}^e \delta(\Omega^2 - \Omega_e{}^2)\}. \tag{18a}$$

Here

$$\Omega_{zk}{}^f = \Omega_{kz}{}^f = \Omega_{xy}{}^e = \Omega_{kz}{}^e = 0, \quad \Omega_{xy}{}^f = \Omega_{yx}{}^{*f} = \Omega_{xy}{}^e = \Omega_{yx}{}^{*e} = i\Omega, \quad \Omega_{xx}{}^f = \Omega_1{}^f, \quad \Omega_{yy}{}^f = \Omega_2{}^f,$$

$$\Omega_f{}^2 = \Omega_1{}^f \Omega_2{}^f, \quad \Omega_2{}^f - \Omega_1{}^f = 4\pi\gamma^+ M \sin^2(\widehat{\mathbf{q}\mathbf{M}});$$

$$\Omega_1{}^f = \gamma^+ \left[H_0 + \frac{1}{2}({}^1H_A + {}^2H_A) \right]$$ — ferromagnetic resonance frequency

$$\Omega_{xx}{}^e = \Omega_{yy}{}^e = \Omega_e = \lambda(\gamma^+ M + \gamma^- L)$$ — exchange resonance frequency

For an antiferromagnetic ("easy axis" type)

$$\langle \tilde{M}_i \tilde{L}_k \rangle_{\Omega,\ q\to 0} = \langle \tilde{L}_i \tilde{M}_k \rangle_{\Omega,\ q\to 0} = \sqrt{\frac{\lambda L}{H_A}} \langle \tilde{M}_i \tilde{M}_k \rangle_{\Omega,\ q\to 0} = \sqrt{\frac{HA}{\lambda L}} \langle L_i L_k \rangle_{\Omega, q\to 0} = \frac{\pi\hbar\gamma^+ L}{2} \operatorname{cth} \frac{\hbar\Omega}{2T} \times \{\Omega_{ik}{}^+ \delta(\Omega^2 - \Omega_+{}^2) + \Omega_{ik}{}^- \delta(\Omega^2 - \Omega_-{}^2)\}, \tag{18b}$$

where

$$\Omega_{zk}{}^\pm = \Omega_{kz}{}^\pm = 0, \quad \Omega_{xy}{}^\pm = \Omega_{yx}^{*\pm} = i\Omega;$$

$\Omega^{\pm}_{xx} = \Omega^{\pm}_{yy} = \Omega_\pm = \gamma\sqrt{(\lambda L H_A)} \pm \gamma H_0$ are the antiferromagnetic resonance frequencies. In all expressions for the correlators

$$z \parallel \delta = \mathbf{M}/M, \quad y \parallel [\delta \mathbf{q}].$$

[4)]For a ferromagnetic one can obtain the correlators $\langle \tilde{M}_i \tilde{M}_k \rangle$ from (18a), putting $\gamma^- = 0$. We then obtain expressions which by other means were obtained by Akhiezer and Bolotin.[10]

6. ANGULAR CHARACTERISTICS AND SPECTRUM OF RAMAN SCATTERING

We shall assume for the sake of simplicity that the ferromagnetic resonance frequency $\Omega^f > 4\pi\gamma^+ M$. Then $\Omega_1^f = \Omega_2^f = \Omega_f$ and all correlators occurring in the theory of MRS and ERS will have the same simple structure:

$$\langle A_i A_k \rangle_{\Omega, \mathbf{q}} \equiv \langle A^2 \rangle_{|\Omega|, \mathbf{q}} \tau_{ik}(\Omega), \quad \tau_{kz} = 0, \quad \tau_{xx} = \tau_{yy} = 1, \quad \tau_{xy} = i\Omega/|\Omega|. \tag{19}$$

Using only this property of them which is more general than the model assumed by us of the magnetic structure we can obtain the angular characteristics of the scattering. For natural light, e.g., it follows from (16) and (19) that

$$\frac{dh(\omega^{ex} + \Omega)}{d\theta} = [1 - \cos\alpha\beta \cos\alpha\delta \cos\beta\delta] \frac{\varepsilon^2\omega^4}{2c^4} \langle A^2 \rangle_{|\Omega|, 0}. \tag{20a}$$

Here, as usual, α and β are the directions of propagation of the scattered and incident light, and δ the direction of the magnetization. For scattering at right angles $\cos\alpha\beta = 0$ and the extinction coefficient $dh/d\theta$ is independent of the direction of the magnetization.[5)] Integrating (20a), we get

$$h(\omega^{ex} + \Omega) = \left[1 - \frac{\cos^2 \widehat{\beta\delta}}{3}\right] \frac{2\pi\varepsilon^2\omega^4}{c^4} \langle A^2 \rangle_{|\Omega|, 0}. \tag{20b}$$

In the case of scattering of circularly polarized light we have instead of (20b)

$$h^\pm(\omega^{ex} + \Omega) = \left[1 - \frac{\cos^2 \widehat{\beta\delta}}{3} \pm \frac{\Omega}{|\Omega|} \frac{\cos \widehat{\beta\delta}}{3}\right] \frac{2\pi\varepsilon^2\omega^4}{c^4} \langle A^2 \rangle_{|\Omega|, 0}. \tag{20c}$$

The intensity of the Stokes and the anti-Stokes components can thus be essentially different (by a factor three when $\cos\beta\delta = 1$). Moreover, the intensity of the Stokes component when right-handedly polarized light is scattered is the same as the intensity of the anti-Stokes component for left-handedly polarized light, and vice versa. In the case of linearly polarized light the total extinction coefficient h looks the same as for natural light (see (20b)). However, the angular distribution of the intensity depends on the direction of the polarization of the light, and on the size and shape of the sample. For scattering at right angles for a sample, the shape of which has a center of inversion

$$\frac{dh_\varphi}{d\theta} = \frac{\varepsilon^2\omega^4}{2c^4} \langle A^2 \rangle_{|\Omega|, 0} [1 + 2\cos(\widehat{\beta\delta}) \times (\cos\varphi \cos\widehat{\beta\delta} + \sin\varphi \cos\widehat{\alpha\delta})]. \tag{20d}$$

[5)]When the assumption $\Omega^f \gg 4\pi\gamma^+ M$ is no longer satisfied we get in a ferromagnetic for the case $\alpha \perp \beta$

where

$$J = \frac{1}{V}\int_V \cos[2k_0(\mathbf{G}\boldsymbol{\beta})(\mathbf{r}\boldsymbol{\beta})]\,d^3r.$$

Here φ is the angle between the plane of polarization in the center of the sample and the direction $[\boldsymbol{\alpha}\times\boldsymbol{\beta}]$. For a spherical sample of radius R

$$J = \frac{3}{\psi^2}\left[\frac{\sin\psi}{\psi} - \cos\psi\right].$$

Here $\psi = 2Rk_0(\mathbf{G}\cdot\boldsymbol{\beta})$ is the angle of rotation of the polarization plane over a length equal to the diameter of the sample. When $\psi \ll \pi$ this expression is equal to unity and vanishes for the first time when $\psi \approx \pi/6$.

For a parallelepiped with edges l_x, l_y, l_z

$$J = \frac{\sin\psi_x \sin\psi_y \sin\psi_z}{\psi_x\psi_y\psi_z},$$

where $\psi_x = k_0(\mathbf{G}\cdot\boldsymbol{\beta})(\mathbf{l}_x\cdot\boldsymbol{\beta})$ is the angle between the polarization planes of the light at opposite faces (along the direction $\mathbf{l}_x$) of the parallelepiped.

We note that Eqs. (20a, c) refer equally well to ERS; in (20d) it is only necessary to change the sign in front of J.

In conclusion we estimate the intensity of the one-magnon Raman scattering of light. From (10), (18a), and (19) we get for a ferromagnetic:

$$\langle A^2\rangle_{|\Omega|,0} = \left(\frac{G}{2\pi}\right)^2 \frac{\pi\hbar\gamma^+}{2M}\,\mathrm{cth}\frac{\hbar\Omega}{2T}\,\delta(\Omega-\Omega_f). \qquad (21)$$

Substituting this into (20b) and bearing in mind that $\pi\gamma\hbar/2M \sim a^3$ where a is the lattice constant, we get easily the estimates (1a) and (1b).

From (18a) it is clear that an estimate of the quantity $\langle A^2\rangle_{|\Omega|,0}$ for an exchange line of a ferromagnetic will contain an additional small parameter $(\gamma^-/\gamma^+)^2 < 10^{-3}$. In an antiferromagnetic an estimate of $\langle A^2\rangle_{|\Omega|,0}$ also will contain the additional parameter $\sqrt{(H_A/\lambda L)}$ which can be small: $\sqrt{(H_A/H_E)} = \sqrt{(H_A/\lambda L)} \approx 0.5$ to 10^{-2}. Similar estimates can also be made for ERS.

I express my deep gratitude to V. L. Gurevich for posing the problem and supervising my work. It is my pleasant duty to thank A. I. Ansel'm, G. M. Nedlin, A. G. Gurevich, O. V. Konstantinov, and B. I. Shklovskiĭ for fruitful discussions of this ppp paper and useful advice.

[1] F. G. Bass and M. I. Kaganov, Zh. Eksp. Teor. Fiz. 37, 1390 (1959) [Sov. Phys.-JETP 10, 989 (1960)].

[2] R. J. Elliott and R. Loudon, Phys. Letters 3, 189 (1963); Y. R. Shen and N. Bloembergen, Phys. Rev. 143, 372 (1966).

[3] R. A. Fleyru, S. P. S. Porto, L. E. Cheesman, and H. J. Guggenheim, Phys. Rev. Letters 17, 84 (1966).

[4] G. A. Smolenskiĭ, V. M. Yudin, P. P. Syrnikov, and A. B. Sherman, Fiz. Tverd. Tela 8, 2965 (1966); Sov. Phys.-Solid State 8, 2368 (1967).

[5] L. D. Landau and E. M. Lifshitz, Elektrodinamika sploshnykh sred (Electrodynamics of continuous media), Gostekhizdat, 1959, Sec. 60 (English translation published by Pergamon, 1960).

[6] L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Fizmatgiz, 1963, Sec. 114 (English translation published by Pergamon, 1965).

[7] O. V. Konstantinov and V. I. Perel', Zh. Eksp. Teor. Fiz. 37, 786 (1959) [Sov. Phys.-JETP 10, 560 (1960)].

[8] E. A. Turov, Fizicheskie svoĭstva magnitouporyadochennykh kristallov (Physical properties of magnetically ordered crystals) AN SSSR, 1964.

[9] L. D. Landau and E. M. Lifshitz, Statisticheskaya fizika (Statistical Physics), Nauka, 1964, Ch. 12 (English translation published by Pergamon, 1967).

[10] I. A. Akhiezer and Yu. V. Bolotin, K teorii fluktuatsiĭ i rasseyaniya medlennykh neĭtronov v ferromagnetikikh (On the Theory of Fluctuations and the Scattering of Slow Neutrons in Ferromagnetics) Preprint FTI AN USSSR, Khar'kov, 1967.

[11] B. Lax and K. J. Button, Microwave ferrites and ferrimagnetics, McGraw-Hill, 1962.

[12] V. M. Genkin, G. M. Genkin, and V. M. Faĭn, Proc. Second All-Soviet Symposium on Nonlinear Optics, Izd. SO AN SSSR, Novosibirsk, 1966.

Translated by D. ter Haar
19