# *RAMAN SCATTERING OF ELECTROMAGNETIC WAVES IN FERROMAGNETIC DIELECTRICS*

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A phenomenon of Raman scattering of electromagnetic waves by fluctuations of the magnetic moment is predicted. The coefficient of extinction for the scattered radiation is calculated.

RAMAN scattering — that is, the appearance of shifted frequencies in the spectrum of the scattered radiation — is connected with an interaction of the incident electromagnetic wave with the characteristic oscillations of the scattering object. A ferromagnetic dielectric possesses an additional branch of oscillations, produced by propagation of spin waves. The magnetic field of the incident wave interacts with the oscillations of the magnetic moment (with the spin wave), and this also should lead to characteristic Raman scattering. The present communication is devoted to the theoretical elucidation of this phenomenon.

We consider the case in which the frequency $\omega_0$ of the incident wave is much larger than the characteristic frequencies of the ferrodielectric. The discussion applies to waves in the millimeter range or in the far infrared part of the spectrum, since a ferromagnetic dielectric is transparent at these frequencies. It is true, of course, that this can be the case only at sufficiently low temperatures, where the semiconducting properties of the ferrites are not exhibited. Then the system of equations that describes the scattered field in the medium has the form

$$\mathrm{curl}\,\mathbf{H}_{\mathrm{sc}}-\frac{\varepsilon}{c}\frac{\partial\mathbf{E}_{\mathrm{sc}}}{\partial t}=0,\qquad \mathrm{curl}\,\mathbf{E}_{\mathrm{sc}}+\frac{1}{c}\frac{\partial\mathbf{B}_{\mathrm{sc}}}{\partial t}=0,$$

$$\mathbf{B}_{\mathrm{sc}}=\mathbf{H}_{\mathrm{sc}}+4\pi\mathbf{M}_{\mathrm{sc}},\quad \partial\mathbf{M}_{\mathrm{sc}}/\partial t=g\,[\mathbf{M}_{\mathrm{sp}}\times\mathbf{H}_{\mathrm{inc}}].\tag{1}$$

Quantities related to the scattered wave are distinguished by the index "sc"; $\mathbf{M}_{\mathrm{sp}}$ is the oscillating part of the magnetic moment, due to the spin wave; $\mathbf{H}_{\mathrm{inc}}$ is the magnetic field intensity of the incident wave. $\mathbf{H}_{\mathrm{inc}}$ satisfies the linear Maxwell equations for a ferromagnetic dielectric.

Since by hypothesis the frequency of the incident wave is large, the magnetic permeability of the ferrite can be set equal to unity, i.e., $\mathbf{M}_{\mathrm{inc}}=0$. This has permitted omission, in the equation for $\mathbf{M}_{\mathrm{sc}}$, of all terms containing $\mathbf{M}_{\mathrm{inc}}$. Omitted also are terms $g\,[\mathbf{H}_{\mathrm{eff}}\times\mathbf{M}_{\mathrm{sc}}]$, with $\mathbf{H}_{\mathrm{eff}}=\mathbf{H}_0+\beta\mathbf{M}_0$ ($\mathbf{H}_0$ is a constant field applied to the ferrite along its axis of easiest magnetization; $\mathbf{M}_0$ is the saturation magnetic moment; $\beta$ is the anisotropy constant), since by hypothesis $\omega_0\gg gH_{\mathrm{eff}}$.

From the system (1) we have

$$\Delta\mathbf{E}_{\mathrm{sc}}-\frac{\varepsilon}{c^2}\frac{\partial^2\mathbf{E}_{\mathrm{sc}}}{\partial t^2}=\frac{4\pi g}{c}\,\mathrm{curl}\,[\mathbf{M}_{\mathrm{sp}}\times\mathbf{H}_{\mathrm{inc}}].\tag{2}$$

We expand $\mathbf{M}_{\mathrm{sp}}$ as a Fourier integral in time:

$$\mathbf{M}_{\mathrm{sp}}=\int_{-\infty}^{\infty}\mathbf{M}_{\Omega}e^{-i\Omega t}\,d\Omega.\tag{3}$$

Since the magnetic field of the incident wave is proportional to $e^{-i\omega_0 t}$ ($\mathbf{H}_{\mathrm{inc}}=\mathbf{H}_{\omega_0}e^{-i\omega_0 t}$), we get from (2) for the Fourier component $\mathbf{E}_{\omega}$ of the scattered field, where $\omega=\omega_0+\Omega$,

$$\mathbf{E}_{\omega}(\mathbf{r})=-\frac{g}{c}\int\mathrm{curl}'\,[\mathbf{M}_{\Omega}(\mathbf{r}')\times H_{\omega_0}(\mathbf{r}')]\,\frac{e^{ikR}}{R}\,dv'.\tag{4}$$

where

$$k^2=\omega^2\varepsilon(\omega)/c^2,\qquad R=|\mathbf{r}-\mathbf{r}'|.\tag{5}$$

On taking into account that the distance to the point at which the field is sought is much larger than the linear dimensions of the scattering body (the ferrite), we find from (4)

$$\mathbf{E}_{\omega}=-g\frac{e^{ikR_0}}{R_0c}\int\mathrm{curl}\,[\mathbf{M}_{\Omega}(\mathbf{r}')\times\mathbf{H}_{\omega_0}(\mathbf{r}')]\,e^{-i\mathbf{k}\mathbf{r}'}\,dv',\ \mathbf{k}=k\mathbf{r}/r.\tag{6}$$

Since $\mathbf{H}_{\omega_0}(\mathbf{r}')=\mathbf{H}_{\omega_0}e^{i\mathbf{k}_0\mathbf{r}'}$, where $k_0^2=\omega_0^2\epsilon(\omega_0)/c^2$, it is easy to obtain from the last expression

$$E_{\omega}=-ig\frac{e^{ikR_0}}{cR_0}\int\{\mathbf{M}_{\Omega}(\mathbf{r}')(\mathbf{k}\mathbf{H}_{\omega_0})-\mathbf{H}_{\omega_0}(\mathbf{k}\mathbf{M}_{\Omega}(\mathbf{r}'))\}\,e^{-i\mathbf{q}\mathbf{r}'}\,dv',\tag{7}$$

where $\mathbf{q}=\mathbf{k}-\mathbf{k}_0$.

We are interested in the scattering of an electromagnetic wave by thermal fluctuations of magnetic moment; that is, by spin waves of random amplitude. Naturally the mean value of the scattered field in this case is zero, since $\langle M_\Omega \rangle = 0$. Here and hereafter, $\langle f \rangle$ denotes the statistical mean of the quantity f.

For description of the properties of the scattered radiation, it is convenient to introduce the tensor

$$I_{ik} = \langle E_i E_k \rangle_\omega, \tag{8}$$

which according to (6) is equal to

$$I_{\alpha\beta} = g^2 \frac{k^2 V}{R_0^2 c^2} \Big\{ H_z^2 \int \langle M_\alpha M_\beta \rangle_\Omega e^{-iq\rho}\, dv_\rho - H_\beta H_z \int \langle M_\alpha M_z \rangle_\Omega e^{-iq\rho} dv_\rho - H_\alpha H_z \int \langle M_\beta M_z \rangle_\Omega e^{-iq\rho} dv_\rho + H_\alpha H_z \int \langle M_z M_z \rangle_\Omega e^{-iq\rho}\, dv_\rho \Big\} \qquad (\alpha, \beta = x, y) \tag{9}$$

(V is the volume of the scattering body; $H_i = (H_{\omega_0})_i$).

In obtaining the last formula we have made use of the facts that: (1) in the tensor $\mathbf{I}_{ik}$ only the x and y components differ from zero, if the z axis is taken along the vector $\mathbf{k}$; (2) $\langle M_i(\mathbf{r}) M_k(\mathbf{r}') \rangle_\Omega \equiv \langle M_i M_k \rangle_\Omega$ depends only on $\rho = \mathbf{r} - \mathbf{r}'$.

It is clear from formula (9) that the tensor components $I_{\alpha\beta}$ are determined by the Fourier transforms $\langle M_i M_k \rangle_{\Omega,q}$ (because of the smallness of the correlation distance as compared with the dimensions of the body, the integration in (9) is extended over all space). If we neglect spatial dispersion of the magnetic susceptibility, then by proceeding as was done, for example, in reference 1, one can obtain

$$\langle M_i M_k \rangle_{\Omega,q} = \langle M_i M_k \rangle_{\Omega,0} = \frac{\hbar}{2\pi} \chi''_{ik}(\Omega) \coth \frac{\hbar\Omega}{2T},$$

$$\chi''_{ik}(\Omega) = (1/2i)\{\chi_{ik}(\Omega) - \chi^*_{ki}(\Omega)\}. \tag{10}$$

Here $\chi_{ik}(\Omega)$ is the magnetic susceptibility tensor of the ferrite.

Neglect of spatial dispersion is legitimate if the incident wavelength satisfies the following condition:

$$\lambda \gg a \sqrt{I / g\hbar H_{\text{eff}}} \sim (30 \text{ to } 100)\, a, \tag{11}$$

where I is the exchange integral ($I \sim 10^{-13}$ erg) and a is the interatomic distance. This condition shows that up to ultraviolet frequencies the incident wave interacts chiefly with the uniform oscillation of the magnetic moment. From (10) and (9) we have

$$I_{\alpha\beta} = g^2 \frac{k^2 \hbar H^2 V}{2\pi R_0^2 C^2} \coth \frac{\hbar\Omega}{2T} \{ e_z^2 \chi''_{\alpha\beta} - e_\beta e_z \chi''_{\alpha z} - e_\alpha e_z \chi''_{\beta z} + e_\alpha e_\beta \chi''_{zz} \}. \tag{12}$$

We assume that the incident wave is linearly polarized. The vector $\mathbf{e} = \mathbf{H}/H$ is its polarization unit-vector.

By definition, the extinction coefficient h is equal to

$$h = (R_0^2 / V H^2) \iint I_{\alpha\alpha}(\Omega)\, do\, d\omega, \qquad \omega = \omega_0 + \Omega, \tag{13}$$

where do is an element of solid angle (the integration is extended over all directions of the wave vector of the scattered wave). On substituting (12) in (13) and carrying out the integration, we get

$$h = \frac{4}{3} g^2 \frac{k^2 \hbar}{c^2} \int \coth \frac{\hbar\Omega}{2T} \{ \chi''_{jj} - (\mathbf{e}, \hat{\chi}'' \mathbf{e}) \}\, d\omega, \tag{14}$$

where $\hat{\chi}''\mathbf{e}$ is the vector with components $\chi''_{ik} e_k$.

If $\chi''_{ik} = \chi'' \delta_{ik}$, that is, if the anisotropy of the tensor $\chi''_{ik}$ can be neglected, then

$$h = (8 g^2 k^2 \hbar / 3c^2) \int \chi''(\Omega) \coth(\hbar\Omega / 2T)\, d\Omega. \tag{15}$$

If the incident radiation is unpolarized, then the expression (14) must be averaged over all orientations of the vector $\mathbf{e}$. The result of the averaging is

$$h = 2 g^2 \frac{k^2 \hbar}{c^2} \left(1 - \frac{1}{3} \cos^2 \theta\right) \int \chi''_\perp \coth \frac{\hbar\Omega}{2T}\, d\omega. \tag{16}$$

In the averaging we assumed that $\chi''_{11} = \chi''_{22} = \chi''_\perp$ and that $\chi_{j3} = \chi_{3j} = 0$ for $j = 1, 2, 3$; $\theta$ is the angle between axis 3 and the direction of propagation of the scattered wave, $\mathbf{k}_0/k_0$.

The tensor $\chi''_{ik}$ that was used is characteristic of ferrites (with axis 3 pointed along an axis of easiest magnetization) if we suppose that we have to do with a ferrite magnetized to saturation. Under these conditions, the magnetic susceptibility is determined by rotation of the magnetic moment under the influence of the external field.

The expression (16) determines the total intensity (at all frequencies) of the scattered radiation. The integrand is the density of radiation in the frequency interval $(\omega, \omega + d\omega)$. Since $\chi_\perp(\omega)$ has a resonance character, the scattered radiation is concentrated near the frequencies $\omega = \omega_0 \pm \omega_r$, where $\omega_r = gH_{\text{eff}}$ is the ferromagnetic resonance frequency:*

$$h = \int I_\omega \delta(\omega \pm \omega_r)\, d\omega,$$

$$I_\omega = \pi (1 - {}^1\!/_3 \cos^2 \theta)\, g^3 (k^2 \hbar M / c^2) \coth(\hbar\omega_r / 2T). \tag{17}$$

In the derivation of the last formula, we used

*$\omega_r = gH_{\text{eff}}$ is the frequency of uniform ferromagnetic resonance for an infinite ferrite or a specimen in the form of a sphere. For specimens of more complicated shape, the resonance frequency depends on the demagnetizing factors. The possibility of neglecting the shape of the specimen is based on the fact that representative dimensions L of the specimen are large ($L \gg c/\omega_r \sim 1$ cm).

the expression for $\chi''_\perp(\omega)$ known from the theory of ferromagnetic resonance:

$$\chi''_\perp(\Omega) = \lim_{\eta\to 0} \frac{1}{4}\eta \frac{gM\omega_r}{(\omega_r \pm \Omega)^2 + \eta^2\omega_r^2}. \quad (18)$$

Here we have neglected the finite width of the ferromagnetic resonance line. If we take account of the line width, then according to (16) and (18) we get

$$h = \frac{1}{2}\left(1 - \frac{1}{3}\cos^2\theta\right)\frac{g^3k^2\hbar M\omega_r}{c^2}\eta\int_0^\infty \frac{\operatorname{curl}(\hbar\Omega/2T)\,d\Omega}{(\Omega \pm \omega_r)^2 + \eta^2\omega_r^2}, \quad (19)$$

where $\eta = \lambda/gM$; $\lambda$ is the relaxation constant in the equation of motion of the magnetic moment.

In order to estimate the magnitude of the extinction coefficient, we may use formula (17). If we assume $\hbar\omega_r \ll 2T$ and take the incident wavelength equal to 1 mm, then $h \approx 10^{-16}$. Such extinction coefficients are easy to measure by modern methods of radio technology. Study of the dependence of $I_\omega$ on frequency provides a possibility of determining the resonance frequencies (and line shapes) of a ferromagnetic dielectric.

In closing, we mention that the radiation scattered by a ferrite possesses peculiar polarization properties. Because of the gyrotropy of the scattering medium, the scattered radiation is a mixture (superposition) of two circularly polarized beams, with the relative intensity different in different directions.

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[1] L. D. Landau and E. M. Lifshitz, Электродинамика сплошных сред (Electrodynamics of Continuous Media), Fizmatgiz, 1958.

Translated by W. F. Brown, Jr.
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