Theory of nonlinear processes in paramagnetic crystals

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The kinetic equations for reciprocal temperatures of the Zeeman and dipole-dipole reservoirs are obtained by solving the Liouville equation to within terms of third order in the interaction with an alternating magnetic field. It is shown that in an alternating magnetic field whose amplitude is much less than the local magnetic field the results obtained are identical with those given by the Provotorov theory {Zh. Eksp. Teor. Fiz. **41**, 1582 (1961), **42**, 882 (1962) [Sov. Phys.-JETP **14**, 1126 (1962), **15**, 611 (1962)] etc.}, whereas in the case of a strong alternating field they are identical with the results of the Redfield theory [Phys. Rev. **98**, 1787 (1955)]. The phenomenon of second harmonic generation in a paramagnetic crystal is considered theoretically allowing for the dipole-dipole interaction temperature.

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The dyanmic behavior of the electron and nuclear spin systems in a solid subjected to an alternating magnetic field is being described successfully in the current literature on the basis of the two-temperature Provotorov theory.[1–3] The kinetic equations for the reciprocal temperatures of the Zeeman and dipole–dipole reservoirs are derived in[1,2] using a form of the density matrix found by solving the Liouville equation to within terms of second order in the operator $\mathscr{H}_t$ describing the interaction of the spin system with the alternating magnetic field. Therefore, the Provotorov theory is applicable when an alternating magnetic field H_1 is much less than the amplitude of the local magnetic field H_L. The problem of saturation of paramagnetic resonance by a strong electromagnetic field ($H_1 \gtrsim H_L$) was solved by Redfield[4] using the representation of a single spin temperature in a rotating coordinate system. It is known[5] that the transition to a rotating coordinate system is equivalent to the retention of all the terms in the expansion of the density matrix in terms of the operator $\mathscr{H}_t$.

In the first section of the present paper we shall derive the kinetic equations for the reciprocal temperatures including terms of the fourth order in the operator $\mathscr{H}_t$ and applying the Kubo–Tomita method[6] in which the form of the function is determined using the first three terms in a power-series expansion. The equations obtained and their steady-state solutions apply for any relationship between alternating and local magnetic fields. If $H_1 \ll H_L$, the results agree with those of the Provotorov theory[1–3] whereas in the $H_1 \gtrsim H_L$ case, they agree with the Redfield results.[4]

In the succeeding sections we shall discuss the problem of second harmonic generation in paramagnetic crystals. This phenomenon is considered in[5,7–9] but a consistent allowance is not made for the role of the temperature of the dipole–dipole interactions.

1. KINETIC EQUATIONS FOR RECIPROCAL TEMPERATURES

We shall consider a paramagnetic crystal subjected to a static magnetic field $\mathbf{H}_0$ parallel to the z axis. An alternating magnetic field $\mathbf{H}_1$ of frequency ω is applied at an angle θ relative to the static field. The density matrix of this system satisfies the equation

$$\frac{\partial}{\partial t}\rho(t)=\frac{1}{i\hbar}[Z+\mathscr{H}_d{}^0+\mathscr{H}_d{}'+L+\mathscr{H}_t+G,\ \rho(t)],\tag{1}$$

where Z is the Zeeman energy of the spin system in the static field:

$$Z=-\gamma\hbar H_0 I_z=\hbar\omega_0 I_z,\tag{2}$$

$\mathscr{H}_d^0$ and $\mathscr{H}_d'$ are the secular and nonsecular parts of the dipole–dipole interaction, respectively; L is the Hamiltonian of the lattice; G is the Hamiltonian of the spin–lattice interaction; $\mathscr{H}_t$ is the Hamiltonian of the interaction of the spin system with the alternating magnetic field:

$$\mathscr{H}_t=-2\gamma\hbar(H_{1x}I_x+H_{1z}I_z)\cos\omega t,\tag{3}$$

where $H_{1x}=H_1\sin\theta$, $H_{1z}=H_1\cos\theta$.

If we represent the total Hamiltonian of the system as a sum of two operators

$$\mathscr{H}_0=Z+L+\mathscr{H}_d{}^0,\quad V_t=\mathscr{H}_d{}'+G+\mathscr{H}_t\tag{4}$$

and then use the interaction representation, we find the solution of Eq. (1) in the form

$$\rho(t)=\rho_0(t)+\rho_1(t)+\rho_2(t)+\ldots+\rho_n(t),\tag{5}$$

where

$$\rho_n(t)=\left(\frac{1}{i\hbar}\right)^n\int\limits_{t'-t}^{0}d\tau_1\int\limits_{t'-t}^{\tau_1}d\tau_2$$

$$\ldots\int\limits_{t'-t}^{\tau_{n-1}}d\tau_n[\tilde{V}_{t+\tau_1}(\tau_1),\ [\tilde{V}_{t+\tau_2}(\tau_2),\ldots,\ [\tilde{V}_{t+\tau_n}(\tau_n),\rho_0(t)]\ldots],\tag{6}$$

$$\tilde{V}_{t+\tau_n}(\tau_n)=\exp\left\{\frac{i}{\hbar}\mathscr{H}_0\tau_n\right\}(\mathscr{H}_d{}'+G+\mathscr{H}_{t+\tau_n})\exp\left\{-\frac{i}{\hbar}\mathscr{H}_0\tau_n\right\},\tag{7}$$

$$\rho_0(t)=\exp\left\{-\frac{i}{\hbar}\mathscr{H}_0(t-t')\right\}\rho(t')\exp\left\{\frac{i}{\hbar}\mathscr{H}_0(t-t')\right\}.\tag{8}$$

It should be noted that $\rho_0(t)$ describes such a state of a paramagnetic sample in which it undergoes a transition from a state with a density matrix $\rho(t')$ in a time $t-t'$ on condition that the total Hamiltonian of the sample is

$\mathscr{H}_0$. It is known[1-3] that as a result of evolution in a time of the order of the spin-spin relaxation time T_2 a system of paramagnetic particles described by the Hamiltonian $\mathscr{H}_0$ reaches a quasiequilibrium state with two different temperatures differing from the lattice temperature. Therefore, when $t-t' \gtrsim T_2$, the high-temperature approximation gives

$$\rho_0(t)=\frac{\exp\{\alpha(t)Z+\beta(t)\mathscr{H}_d{}^0-L/kT\}}{\mathrm{Tr}\,\exp\{\alpha(t)Z+\beta(t)\mathscr{H}_d{}^0-L/kT\}}$$
$$=(1-\alpha(t)Z-\beta(t)\mathscr{H}_d{}^0)\frac{\exp\{-L/kT\}}{(2I+1)^N\,\mathrm{Tr}\,\exp\{-L/kT\}}, \tag{9}$$

where N is the total number of paramagnetic particles in a sample; $\alpha(t)$ and $\beta(t)$ are the reciprocal temperatures of the Zeeman and dipole–dipole reservoirs, respectively; T is the lattice temperature (assumed to be constant); and k is the Boltzmann constant. If $t-t' \gtrsim T_2$, the lower limit of integration in Eq. (6) can be extended to $-\infty$.[2,10]

The kinetic equations for the reciprocal temperatures are obtained from

$$\mathrm{Tr}\,Z\frac{\partial}{\partial t}\rho(t)=\frac{1}{i\hbar}\,\mathrm{Tr}\,Z[\mathscr{H}_d{}'+G+\mathscr{H}_t,\rho(t)], \tag{10}$$

$$\mathrm{Tr}\,\mathscr{H}_d{}^0\frac{\partial}{\partial t}\rho(t)=\frac{1}{i\hbar}\,\mathrm{Tr}\,\mathscr{H}_d{}^0[\mathscr{H}_d{}'+G+\mathscr{H}_t,\rho(t)]. \tag{11}$$

This is done by substituting in Eqs. (10) and (11) the values of $\rho(t)$ calculated from Eq. (5) to within the third approximation.

We shall first write the term which is quadratic in the operator $\mathscr{H}_t$ and which appears on the right-hand-side of Eq. (10) after the substitution of $\rho_1(t)$:

$$-\frac{i\omega_1{}^2\hbar\omega_0}{2}\int\limits_{-\infty}^{0}d\tau\,\mathrm{Tr}\,I_+[\tilde{I}_-(\tau),\rho_0(t)]e^{i\omega\tau}$$
$$=-\frac{\omega_1{}^2\hbar^2\omega_0{}^2}{2}\left(\alpha-\beta\frac{\omega_0-\omega}{\omega_0}\right)\int\limits_{-\infty}^{\infty}\mathrm{Tr}\,I_+I_-(\tau)\exp[-i(\omega_0-\omega)\tau]d\tau, \tag{12}$$

where $\omega_1=\gamma H_{1x}$, $I_\pm=I_x\pm iI_y$;

$$\left.\begin{aligned}\tilde{I}_\pm(\tau)&=\exp\left\{\frac{i}{\hbar}\mathscr{H}_0\tau\right\}I_\pm\exp\left\{-\frac{i}{\hbar}\mathscr{H}_0\tau\right\}=\exp(\pm i\omega_0\tau)I_\pm(\tau),\\ I_\pm(\tau)&=\exp\left\{\frac{i}{\hbar}\mathscr{H}_d{}^0\tau\right\}I_\pm\exp\left\{-\frac{i}{\hbar}\mathscr{H}_d{}^0\tau\right\}.\end{aligned}\right\} \tag{13}$$

We shall now calculate the trace on the right-hand side of Eq. (12):

$$\mathrm{Tr}\,I_+I_-(\tau)=\mathrm{Tr}\,I_+I_-(1-{}^1\!/_2\tau^2\sigma_0{}^2)\approx\mathrm{Tr}\,I_+I_-\exp\{-{}^1\!/_2\tau^2\sigma_0{}^2\}, \tag{14}$$

where $\sigma_0=\mathrm{Tr}[I_+,\mathscr{H}_d^0][\mathscr{H}_d^0,I_-]/\hbar^2\,\mathrm{Tr}\,I_+I_-$ is the second moment of the absorption curve in a weak rf field.[6] In the calculation of Eq. (14) we shall use the Kubo–Tomita method[6] in which an exponential operator $\exp\{\pm i\mathscr{H}_d^0\tau/\hbar\}$ is expanded as a power series retaining terms up to the second degree of $\sim\hbar^{-n}\langle(\mathscr{H}_d^0)^n\rangle\tau^n$.

After calculation of the spur, the terms of the series are regarded as an expansion of some exponential function in powers of $\sim\hbar^{-n}\langle(\mathscr{H}_d^0)^n\rangle\tau^n$ and the form of this function is reconstructed from the first three terms of the series with $n=0, 1, 2$.

Forestalling the later parts of the paper, we shall mention that the terms quadratic in respect of the operators G and $\mathscr{H}_d'$ appear on the right-hand sides of Eqs. (10) and (11) as a result of substitution of the first approximation from Eq. (5) and they describe the spin–lattice relaxation of the Zeeman and secular dipole–dipole subsystems and also the thermal mixing of these subsystems under the influence of the nonsecular part of the dipole–dipole interaction [see Eq. (23)]. Thus, substituting in Eqs. (10) and (11) only the first two terms $\rho_0(t)+\rho_1(t)$ from Eq. (5), we obtain the kinetic equations of Provotorov for the reciprocal temperatures in the laboratory coordinate system.[1,2]

As a result of substitution of $\rho_3(t)$ in Eq. (10), the right-hand side of this equation acquires a term of the fourth degree in $\mathscr{H}_t$, which—after a change in the order of integration with respect to τ_1, τ_2, and τ_3—becomes

$$2^4 i\cos\omega t\cdot\omega_1{}^2\hbar\omega_0\int\limits_{-\infty}^{0}d\tau_3\int\limits_{\tau_3}^{0}d\tau_2\int\limits_{\tau_2}^{0}d\tau_1\cos\omega(t+\tau_1)\cos\omega(t+\tau_2)\cos\omega(t+\tau_3)$$
$$\times\{\omega_1{}^2\,\mathrm{Tr}\,I_y[\tilde{I}_x(\tau_1),[\tilde{I}_x(\tau_2),[\tilde{I}_x(\tau_3),\rho_0(t)]]]+(\gamma H_{1z})^2\,\mathrm{Tr}\,I_y[\tilde{I}_x(\tau_3),\rho_0(t)]\}. \tag{15}$$

We shall discuss a typical term in Eq. (15):

$$\int\limits_{-\infty}^{0}d\tau_3\int\limits_{\tau_3}^{0}d\tau_2\int\limits_{\tau_2}^{0}d\tau_1\,I_+[\tilde{I}_-(\tau_1),[\tilde{I}_+(\tau_2),[\tilde{I}_-(\tau_3),\rho_0(t)]]]\exp[i\omega(t+\tau_1-\tau_2+\tau_3)]. \tag{16}$$

Expanding the operators $I_\pm(\tau)$ as the Fourier integrals[1,6]

$$I_\pm(\tau)=\int\limits_{-\infty}^{\infty}d\tilde{\omega}\,I_{\tilde{\omega}}^{\pm}e^{i\tilde{\omega}\tau},\quad I_{\tilde{\omega}}^{\pm}=\frac{1}{2\pi}\int\limits_{-\infty}^{\infty}d\tau\,I_\pm(\tau)\,e^{-i\tilde{\omega}\tau}, \tag{17}$$

and using the results in[1,6], we shall represent Eq. (16) in the form

$$2e^{i\omega t}\int\limits_{-\infty}^{0}d\tau_3\int\limits_{\tau_3}^{0}d\tau_2\int\limits_{\tau_2}^{0}d\tau_1\,\mathrm{Tr}\,I_+[\tilde{I}_-(\tau_3),\rho_0(t)]$$
$$\times\exp[-i\tau_1(\omega_0-\omega)]\exp[-i\tau_2(\omega_0-\omega)]\exp[i\omega\tau_3]. \tag{18}$$

It follows from Eqs. (12) and (14) that $\mathrm{Tr}\,I_+[\tilde{I}_-(\tau_3),\rho_0]$ is large in the range $\tau_3\lesssim\sigma_0$, so that near resonance, i.e., when $|\omega_0-\omega|\lesssim\sigma_0$, satisfactory approximation is obtained if Eq. (18) is replaced with

$$e^{i\omega t}\int\limits_{-\infty}^{0}d\tau_3\,\mathrm{Tr}\,I_+[\tilde{I}_-(\tau_3),\rho_0(t)]\tau_3{}^2\exp(i\omega\tau_3). \tag{19}$$

All the terms in Eq. (15) can then be calculated similarly. We shall group on the right-hand side of Eq. (10) the slowly varying terms obtained after the substitution of the first and third approximations:

$$-i\omega_1^2\hbar\omega_0\int_{-\infty}^{\infty} d\tau\,\mathrm{Tr}\,I_+[\tilde{I}_-(\tau),\rho_0]e^{i\omega\tau}\Big\{1-\frac{1}{2}\omega_1^2\tau^2 -\frac{i\omega_1^2\tau}{\omega_0+\omega}-\frac{\omega_1^2}{(\omega_0+\omega)^2}+\sum_{m=\pm1,\pm2}\Big(\frac{i\tau}{m\omega_0}\sigma_{0m}^2-\frac{\sigma_{0m}^2}{m^2\omega_0^2}\Big)\Big\}, \tag{20}$$

where σ_{0m}^2 is given by Eq. (5.12) in[6]. The terms in the braces in Eq. (20) should be regarded as the first terms of the expansion of an exponential function in powers of $\hbar^{-n}\langle(\mathscr{H}_t+\mathscr{H}_d')^n\rangle\tau^n$. Representing the integrand in Eq. (20) as an exponential function of τ and using Eq. (14), we find that integration gives

$$-\Big(1-\sum_m\frac{\sigma_{0m}^2}{m^2\omega_0^2}-\frac{\omega_1^2}{(\omega_0+\omega)^2}\Big)\frac{\omega_1^2\hbar^2\omega_0^2}{2}\Big(\alpha-\beta\frac{\Delta}{\omega_0}\frac{\sigma_0^2}{\sigma_0^2+\omega_1^2}\Big) \times\Big(\frac{2\pi}{\sigma_0^2+\omega_1^2}\Big)^{1/2}\exp\Big\{-\frac{\Delta^2}{2(\sigma_0^2+\omega_1^2)}\Big\}, \tag{21}$$

where

$$\Delta=\omega_0-\omega+\frac{\omega_1^2}{2(\omega_0+\omega)}-\sum_{m=\pm1,\pm2}\frac{\sigma_{0m}^2}{m\omega_0}. \tag{22}$$

Neglecting in Eq. (21) the small (compared with unity) terms $\sigma_{0m}^2/m^2\omega_0^2$ and $\omega_1^2(\omega_0+\omega)^{-2}$, we obtain the final form of the kinetic equations for the reciprocal temperatures:

$$\left.\begin{aligned}\frac{d\alpha}{dt}&=-\Big(\alpha-\frac{\Delta}{\omega_0}\beta\Sigma\Big)\pi\omega_1^2g(\Delta)-(\alpha-\beta)W-\frac{\alpha-\alpha_0}{T_z},\\ \frac{d\beta}{dt}&=\Big(\alpha-\frac{\Delta}{\omega_0}\beta\Sigma\Big)\pi\omega_1^2\frac{\omega_0\Delta}{\omega_L^2}g(\Delta)+(\alpha-\beta)W\frac{\omega_0^2}{\omega_L^2}-\frac{\beta-\beta_0}{T_d},\end{aligned}\right\} \tag{23}$$

where

$$\omega_L^2=\mathrm{Tr}\,(\mathscr{H}_d^0)^2/\mathrm{Tr}\,(I_z)^2,\quad \alpha_0=\beta_0=\frac{1}{kT},\quad \Sigma=\frac{\sigma_0^2}{\sigma_0^2+\omega_1^2},$$

$g(\Delta)$ is a Gaussian curve of half-width $(\sigma_0^2+\omega_1^2)^{1/2}$:

$$g(\Delta)=\frac{1}{[2\pi(\sigma_0^2+\omega_1^2)]^{1/2}}\exp\Big\{-\frac{\Delta^2}{2(\sigma_0^2+\omega_1^2)}\Big\},$$

W is the rate of thermal mixing of the Zeeman and secular dipole–dipole subsystems which is governed by the nonsecular part of the dipole–dipole interaction[1,2]:

$$W=\frac{1}{\hbar^2\,\mathrm{Sp}\,I_z^2}\int_0^\infty \mathrm{Tr}\,[I_z,\mathscr{H}_d'][\tilde{\mathscr{H}}_d(\tau),I_z]d\tau, \tag{24}$$

T_z and T_d are the spin–lattice relaxation times of the Zeeman and dipole–dipole reservoirs, respectively[1,2]:

$$\frac{1}{T_z}=\frac{1}{\hbar^2\,\mathrm{Tr}\,I_z^2\,\mathrm{Tr}\exp\{-L/kT\}}\int_0^\infty d\tau\,\mathrm{Tr}\,[I_z,G][\tilde{G}(\tau),I_ze^{-L/kT}], \tag{25}$$

$$\frac{1}{T_d}=\frac{1}{\hbar^2\,\mathrm{Tr}(\mathscr{H}_d^0)^2\,\mathrm{Tr}\exp\{-L/kT\}}\int_0^\infty d\tau\,\mathrm{Tr}\,[\mathscr{H}_d^0,G][\tilde{G}(\tau),\mathscr{H}_d^0e^{-L/kT}]. \tag{26}$$

It is known from the general theory of nonequilibrium processes[11,12] that the quasiequilibrium density matrix $\rho_0(t)$ in the form given by Eq. (9) can be used to describe a nonequilibrium state if we assume that $\alpha(t)$ and $\beta(t)$ vary slowly with time. Therefore, Eq. (23) is simplified by dropping the rapidly oscillating terms proportional to $\exp(\pm i\omega t)$, $\exp(\pm 2i\omega t)$, etc.

The steady-state solution of the system (23) for $W=0$ is of the form

$$\alpha_{st}=\alpha_0\frac{1+\pi\omega_1^2g(\Delta)T_d\Sigma\Delta^2/\omega_L^2}{1+\pi\omega_1^2g(\Delta)(T_z+T_d\Sigma\Delta^2/\omega_L^2)}, \tag{27}$$

$$\beta_{st}=\beta_0\frac{\pi\omega_1^2g(\Delta)\omega_0\Delta T_d/\omega_L^2}{1+\pi\omega_1^2g(\Delta)(T_z+T_d\Sigma\Delta^2/\omega_L^2)}. \tag{28}$$

If $\omega_1^2\ll\omega_0^2$, i.e., when the Provotorov theory is valid, the system (23) reduces to the Provotorov equations in the laboratory system of coordinates[1,2] and the steady-state solutions of these equations become identical with Eqs. (27) and (28). The formulas (23), (27), and (28) are valid in the range of frequencies near resonance: $|\omega_0-\omega|\lesssim\sigma_0$. Calculations show that when these formulas are used far from resonance ($|\omega_0-\omega|\sim\omega_0$), the parameter $\Sigma=\sigma_0^2/(\sigma_0^2+\omega_1^2)$ in these formulas should be replaced by unity and the half-width of the Gaussian curve $g(\Delta)$ should be taken as equal to σ_0. This is due to the fact that the terms $\sim\tau^2$ originating from Eq. (18) disappear from the integrand in Eq. (20).

2. MACROSCOPIC MAGNETIZATION OF A SAMPLE

We shall calculate the average value of the x component of the magnetization of a sample using

$$\langle M_x\rangle=\mathrm{Tr}\,\rho(t)M_x, \tag{29}$$

where we shall substitute the value of $\rho(t)$ calculated to the fourth approximation $\rho_4(t)$. Applying the results of of[9], we shall express the part of $\langle M_x\rangle$ oscillating at a frequency ω in the form

$$\langle M_x\rangle^\omega=\frac{1}{2}\gamma\hbar^2\omega_1\omega_0\,\mathrm{Tr}\,I_+I_-\Big\{\pi g(\Delta)\Big(\alpha-\beta\frac{\Delta}{\omega_0}\Sigma\Big)\sin\omega t +\Big[\frac{\beta}{\omega_0}\Sigma+\pi g'(\Delta)\Big(\alpha-\beta\frac{\Delta}{\omega_0}\Sigma\Big)\Big]\cos\omega t\Big\}, \tag{30}$$

where $g'(\Delta)$ is defined by

$$g'(\Delta)=-i\Big(\frac{\pi}{2}\Big)^{1/2}g(\Delta)\Phi\Big(\frac{i\Delta}{[2(\sigma_0^2+\omega_1^2)]^{1/2}}\Big), \tag{31}$$

and $\Phi(x)$ is the error function. Substituting in Eq. (30) the steady-state values α_{st} and β_{st}, we obtain the following expressions for the absorption and dispersion signals:

$$v=\frac{\pi\omega_1M_0g(\Delta)}{1+\pi\omega_1^2g(\Delta)(T_z+T_d\Sigma\Delta^2/\omega_L^2)}, \tag{32}$$

$$u=\pi\omega_1M_0\frac{g'(\Delta)+\omega_1g(\Delta)T_d\Sigma\Delta/\omega_L^2}{1+\pi\omega_1^2g(\Delta)(T_z+T_d\Sigma\Delta^2/\omega_L^2)}, \tag{33}$$

where $M_0=\chi_0H_0$ is the equilibrium magnetization of a sample at the lattice temperature.[10] In a strong rf field, when

$$\pi\omega_1^2 g(\Delta)T_Z,\quad \pi\omega_1^2 g(\Delta)T_d \gg 1, \tag{34}$$

bearing in mind that $\sigma_0^2=3\omega_L^2$, we obtain

$$v=\frac{M_0}{\omega_1 3T_d}\frac{\omega_1^2+\sigma_0^2}{\Delta^2+T_Z(\sigma_0^2+\omega_1^2)/3T_d}, \tag{35}$$

$$u=M_0\omega_1\frac{\Delta}{\Delta^2+T_Z(\sigma_0^2+\omega_1^2)/3T_d}. \tag{36}$$

We can easily show that if $\sigma_0^2 \gg \omega_1^2$, the formulas (32), (33), (35), and (36) become identical with the results of the Provotorov theory[1,2] and when the spin-lattice relaxation times are related by $T_z=3T_d$ which is frequently true of solids at low temperatures (see Chap. III in[2]), the formulas (35) and (36) become identical with the Redfield results[4] valid in the $\omega_1^2 \gtrsim \sigma_0^2$ case.

Equation (29) includes also terms oscillating at the doubled frequency:

$$\langle M_x\rangle^{2\omega}=\frac{\pi}{2}\gamma^2\hbar^2 H_{1z}\omega_1\frac{\omega_0}{\omega}\left\{\left(\alpha-\beta\frac{\Delta}{\omega_0}\Sigma\right)[g(\Delta)\sin 2\omega t+g'(\Delta)\cos 2\omega t]\right.$$
$$\left.-\left(\alpha-\beta\frac{\omega_0-\omega}{\omega_0}\right)[f(\omega_0-2\omega)\sin 2\omega t+f'(\omega_0-2\omega)\cos 2\omega t]\right\}. \tag{37}$$

where $f(\omega_0-2\omega)$ is a Gaussian curve of half-width σ_0:

$$f(\omega_0-2\omega)=\frac{1}{(2\pi)^{1/2}\sigma_0}\exp\left\{-\frac{(\omega_0-2\omega)^2}{2\sigma_0^2}\right\}, \tag{38}$$

$$f'(\omega_0-2\omega)=-i\left(\frac{\pi}{2}\right)^{1/2}f(\omega_0-2\omega)\Phi\left(i\frac{\omega_0-2\omega}{2^{1/2}\sigma_0}\right). \tag{39}$$

The second harmonic M_x of the magnetization component has a resonance at $\omega\approx\omega_0$ and $\omega\approx\frac{1}{2}\omega_0$.

We shall use Eq. (29) to calculate the z component of the magnetization, which shows that the second harmonic of the longitudinal magnetization component resonates at $\omega\approx\omega_0$:

$$\langle M_z\rangle^{2\omega}=-\frac{1}{2}\gamma\hbar^2\omega_1^2\frac{\omega_0}{\omega_0+\omega}\operatorname{Tr} I_+I_-\left\{\pi g(\Delta)\left(\alpha-\beta\frac{\Delta}{\omega_0}\Sigma\quad \sin 2\omega t\right.\right.$$
$$\left.+\left[\frac{\beta}{\omega_0}\Sigma+\pi g'(\Delta)\left(\alpha-\beta\frac{\Delta}{\omega_0}\Sigma\right)\right]\cos 2\omega t\right\}. \tag{40}$$

If $\sigma_0^2=3\omega_L^2$ and $T_z=3T_d$, in the case of a strong field (34), (34), the formula (40) is identical with the corresponding expression in[8], where the quantity $\langle M_z\rangle^{2\omega}$ is calculated by a semiclassical method.

3. ELECTROMAGNETIC RADIATION POWER AT DOUBLED FREQUENCY

We shall compare the results obtained with those given in[7] where a study was made of the intensity and phase of electromagnetic radiation at doubled frequency in ruby and diphenyl picryl hydrazyl (DPPH) with different concentrations of paramagnetic atoms. Under the experimental conditions of[7] the radiation power at the frequency 2ω is proportional to

$$[\langle M_x\rangle^{2\omega}\sin\theta+\langle M_z\rangle^{2\omega}\cos\theta]^2.$$

Then, in the frequency range close to $\omega\approx\frac{1}{2}\omega_0$, we find the radiation power

$$P^{2\omega}_{\omega\approx 1/2\omega_0}\sim\left[\frac{\alpha_0 H_1^2\sin^2\theta\cos\theta f(\omega_0-2\omega)}{1+\pi\omega_1^2 f(\omega_0-\omega)(T_Z+T_d\omega_0^2/4\omega_L^2)}\right]^2$$
$$\times\frac{(\omega_0-2\omega)^2+\sigma_0^2}{\sigma_0^2}\cos^2(2\omega t+\varphi), \tag{41}$$

where the phase angle φ is given by the expression

$$\operatorname{tg}\varphi=-\frac{f(\omega_0-2\omega)}{f'(\omega_0-2\omega)}\approx-\frac{\sigma_0}{\omega_0-2\omega}. \tag{42}$$

Here, we have used the representation in the form of the error function series $\Phi(x)\approx 2x/\sqrt{\pi}$ in the range of small x.

Near the frequency $\omega\approx\omega_0$, we obtain

$$P^{2\omega}_{\omega\approx\omega_0}\sim\left[\frac{\alpha_0 H_1^2\sin^2\theta\cos\theta\, g(\Delta)}{1+\pi\omega_1^2 g(\Delta)(T_Z+T_d\Sigma\Delta^2/\omega_L^2)}\right]^2$$
$$\times\left[1+\left(\frac{\Delta}{(\sigma_0^2+\omega_1^2)^{1/2}}-2\Delta T_d\Sigma\omega_1^2/\omega_L^2\right)^2\right]\cos^2(2\omega t+\varphi_1), \tag{43}$$

where

$$\operatorname{tg}\varphi_1=-\frac{g(\Delta)}{g'(\Delta)-2\omega_1^2 g(\Delta)T_d\Sigma\Delta/\omega_L^2}. \tag{44}$$

In weak fields, i.e., when $\omega_1^2 T_z\omega_L^{-1}\ll 1$, we have

$$\operatorname{tg}\varphi_1=-\sigma_0/(\omega_0-\omega), \tag{45}$$

whereas in strong fields, i.e., when the condition (34) is satisfied,

$$\operatorname{tg}\varphi_1=\frac{1}{2\Delta 3T_d}\left(\frac{\sigma_0^2}{\omega_1^2}+1\right). \tag{46}$$

For $\sigma_0^2=3\omega_L^2$ and $T_z=3T_d$, Eq. (46) is identical with the corresponding expression in[8] with the exception of the factor 2 in the denominator. (The absence of the factor 2 in the formula for φ_1 in[8] is due to the fact that the component $\langle M_x\rangle^{2\omega}$ is ignored in[8].) It is clear from Eqs. (42) and (45) that the nature of the dependence of the phase angle on ω in the absence of saturation is the same: the phase angle increases with increasing ω. In the case of saturation at frequencies $\omega\approx\omega_0$, the phase angle increases with decreasing ω, as found experimentally.[7] Moreover, under rf saturation conditions the phase angle φ_1 depends on the alternating magnetic field intensity H_1 and this dependence differs from that used in[7] to explain the experimental results.

The dependence of the second harmonic signal on the frequency of the alternating magnetic field ω is given by Eqs. (41) and (43). In the absence of saturation, the signal is a bell-shaped curve and in the presence of saturation there is a dip at the line center (at $\omega \approx \omega_0$), the depth of the dip depending on the field amplitude H_1. This is exactly the shape of the second harmonic signal which was observed in[7].

There were three factors in[7] which were ignored in our discussion and which could give rise to some discrepancies between the theory and experiment: 1) the field H_1 was applied in the form of pulses and, therefore, was not strictly speaking monochromatic; 2) we ignored the exchange interaction whose magnitude, for certain concentrations of the paramagnetic atoms, could be comparable with or even greater than the magnitude of the dipole–dipole interaction; 3) we ignored the inhomogeneous line broadening.

In conclusion, we shall point out that the functions $g(\Delta)$ and $g'(\Delta)$, as well as the functions $f(\omega_0 - 2\omega)$ and $f'(\omega_0 - 2\omega)$, satisfy the Kramers–Kronig relationships[10] only approximately and only in the frequency range defined by $|\Delta| < (\sigma_0^2 + \omega_1^2)^{1/2}$ and $|\omega_0 - 2\omega| < \sigma_0$. This is due to the fact that the nature of these functions was reconstructed from the first three terms of their expansion as a power series in the operator V_t. One may expect that the retention of the higher terms in the expansion (5) will make it possible to describe higher-order nonlinear effects and to find more accurately the line profiles of the first and second harmonics by analogy with the refinement of the linear response profile obtained by inclusion of terms with higher powers of the dipole–dipole interaction operator.[13]

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Domain structures of {111} iron garnet crystalline plates

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The results are given of an investigation of the domain structure of {111} single-crystal iron garnet plates with a natural tetraxial magnetocrystalline anisotropy. The domain structure was investigated with the aid of the Faraday effect. It was found that the structure was composed of three systems of through stripe domains oriented mainly along the ⟨112⟩ axis and forming a macrodomain structure. Several special features were observed in the behavior of this domain structure in a magnetic field, which was directed at right-angles or parallel to the plate. In particular, macrodomains without an internal substructure were observed during magnetization. The cause of the existence of an unusual hysteresis loop was established. Conditions were found under which the observed pattern became colored (the domains with different magnetization orientations had different "colors"). This effect was used in the identification of magnetic phases. A model of a complex domain structure was developed: it consisted of stripe domains with 71° walls and rhomboidal macrodomains with 109° walls, and the magnetization vectors of the domains were oriented along the ⟨111⟩ axes. The model described satisfactorily the magnetic-field-induced changes in the domain structure. The theoretically calculated relationships between the parameters of the macrodomain structure and stripe substructure were in agreement with the experimental results.

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New practical applications of magnetic domains demand a better understanding of the domain structure of ferromagnets and ferrimagnets (ferrites). The domain structure of magnetically uniaxial materials has been investigated in detail both theoretically and experimentally, but much less work has been done on multiaxial crystals because of the greater variety of domain configurations which can occur in them. This applies particularly to the domain structure of magnetically multiaxial crystals with external surfaces which do not