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[1] L. Cooper, Phys. Rev. **104**, 1189 (1956).

[2] Bardeen, Cooper and Schrieffer, Phys. Rev. **106**, 162 (1957).

[3] Bardeen, Cooper and Schrieffer, Phys. Rev. **108**, 5 (1957).

[4] N. N. Bogoliubov, J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 58 (1958); Soviet Phys. JETP **7**, 41 (1958).

[5] L. D. Landau, J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 262 (1958); Soviet Phys. JETP **7**, 182 (1958).

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138

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THE THERMODYNAMICAL THEORY OF RESONANCE AND RELAXATION PHENOMENA IN FERROMAGNETICS

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The methods of irreversible thermodynamics are applied to derive the time variation of magnetization of ferromagnetics. The role of spin-lattice relaxation in the phanomenon of ferromagnetic resonance is discussed. The resultant equations are compared with those of Landau-Lifshitz and Bloch.

1. In the observation of ferromagnetic resonance, the ferromagnetic specimen is placed in a constant magnetic field $H_0 = H_z$. This magnetizes the sample to saturation. A radiofrequency field **h** is then applied perpendicular to $\mathbf{H}_0$. The amplitude of the field **h** is usually taken to be small $(h \ll H_0)$; therefore, the magnetization vector **M** differs only slightly in direction from $\mathbf{H}_0$. In experiments on the study of relaxation in perpendicular fields, a strong radiofrequency field of high amplitude was applied. This produced a significant deviation of **M** away from $\mathbf{H}_0$.

For the determination of the frequency dependence of the components of the magnetization M_x, M_y, M_z of the ferromagnetic, there are used the equations of Landau-Lifshitz[1], Bloch,[2] or various modifications of these equations,[3] which are frequently put together without sufficient basis.

In the present paper, it is shown that as a result of the application of irreversible thermodynamics, one can obtain (under very simple and general assumptions) equations for the change in the magnetization with time, with consideration both of spin-spin and spin-lattice relaxations, and the role of the latter in the phenomenon of ferromagnetic resonance can also be judged.

From the viewpoint of thermodynamics, we can divide the system of spin moments, which correspond to the magnetic properties of the ferromagnetics, into a separate subsystem with temperature T (the spin system). We shall consider the remaining degrees of freedom of the entire system [analogously to what was done in the thermodynamic theory of paramagnetic relaxation[4]] to be thermostatted, the temperature of which (T_0) we shall consider fixed in the current research. We can show that the latter assumption is related to the conclusions made below and it is easily based on them.

If the subset is found in thermal equilibrium with the thermostat or is isolated completely from it, and the magnetization **M** has a non-equilibrium value, which does not correspond to the field **H**, then we shall call the process of the approximation of M to the equilibrium value the internal or spin-spin relaxation (correspondingly isothermal or adiabatic).

Another form of relaxation will take place if the magnetization of the subsystem M has a constant equilibrium value M_0 while its temperature $T > T_0$. In this case the system will go over to the equilibrium state by a transfer of heat to the thermostat. In what follows, we shall call this process the external or spin-lattice relaxation. Processes of internal and external relaxation usually take place simultaneously and are connected with one another.

The thermodynamic theory of relaxational phenomena, which takes into account both forms of relaxation, was developed in the works of Shaposhnikov and was applied to paramagnetics.[5-7]

2. For a sufficiently rapid change in the field H, the subsystem of spin moments will be found in a non-equilibrium state. The temperature T of the subsystem and the magnetization M do not satisfy the equation of state, which in this case determines a certain value of the field $\mathbf{H}^*$

$$\mathbf{H}^* = \mathbf{H}^*(T, \mathbf{M}), \tag{1}$$

which differs from H. The difference $\mathbf{H}^* - \mathbf{H}$ can be regarded as some additional magnetic field, in the presence of which the subsystem would be in an equilibrium state. By the definition of the field $\mathbf{H}^*$, we have, always,

$$[\mathbf{M}\mathbf{H}^*] = 0. \tag{2}$$

In order to take into account the internal relaxation, we write down the expression for the change in the entropy of the non-equilibrium state of the subsystem, which can be represented in the form (see Ref. 8)

$$TdS = dU - \mathbf{H}d\mathbf{M} + (\mathbf{H} - \mathbf{H}^*)\,d\mathbf{M},$$

where U is the internal energy of the subsystem. The first two terms in the right hand side of the latter equation determine the equilibrium part of the entropy change:

$$T\,(dS)_p = dU - \mathbf{H}d\mathbf{M}, \tag{3}$$

while the latter gives the non-equilibrium part. Therefore, for the measurement of the entropy in the subsystem, we find

$$Td\Delta S/dt = (\mathbf{H} - \mathbf{H}^*)\,d\mathbf{M}/dt.$$

In the approximation of irreversible thermodynamics,[9] the components of the "current" M_k are linear functions of the components of the "force" $(H_k - H_k^*)$, i.e.,

$$\dot{M}_k = \sum_{i=1}^{3} L_{ik}(H_i - H_i^*), \tag{4}$$

where L_{ik} is the tensor of kinetic coefficients whose components are functions of the magnetization for ferromagnetics. Symmetrizing L_{ik}^* and antisymmetrizing L_{ik}^a the parts of the tensor L_{ik} satisfy the Onsager relations:

$$L_{ik}^s(\mathbf{M}) = L_{ik}^s(-\mathbf{M});\ L_{ik}^a(\mathbf{M}) = -L_{ik}^a(-\mathbf{M}). \tag{5}$$

For a magneto-isotropic ferromagnetic, placed in a field $H_0 = H_z$, one should, generally speaking, assume the presence of an axial anisotropy; therefore

$$L_{ik}^s = \lambda_i\delta_{ik}, \quad i, k = 1, 2, 3, \tag{6}$$

where $\lambda_1 = \lambda_2 = \lambda_\perp$, $\lambda_3 = \lambda_\parallel$.

The components of the antisymmetric part of the tensor L_{ik}^a form an axial vector L which, according to (5), is an odd function of the magnetization. The latter requirement can be satisfied by assuming

$$\mathbf{L} = \gamma\mathbf{M}, \tag{7}$$

where the coefficient γ can depend on the temperature.

Now Eq. (4) can be written in the form

$$\dot{M}_k = \lambda_k\{H_k - H_k^*(T, \mathbf{M})\} + \gamma[\mathbf{M}\mathbf{H}]_k. \tag{8}$$

Here the term in curly brackets defines the relaxation process in the spin system. If we neglect this term, then Eq. (8) coincides in form with the equation of motion of a system of noninteracting magnetic momenta in an external magnetic field. In this latter case, the factor γ has the meaning of a magnetomechanical ratio γ_0. In the presence of interaction in the subsystem, γ is generally different fro n γ_0. The dependence of γ on temperature, which is experimentally observed, does not contradict the thermodynamic calculation.

If we do not take the transfer of heat from the spin system to the "lattice" into consideration, then Eq. (8) describes the change in magnetization with time, brought about by the gyroscopic properties of the magnetic momentum and the process of spin-spin relaxation. In order to introduce time relaxation into Eq. (8), we expand the expression for $\mathbf{H}^*(T, \mathbf{M})$ in a series about the equilibrium state of the spin system. Limiting ourselves to the first powers of $\vartheta = T - T_0$ and $\mathbf{m} = \mathbf{M} - \mathbf{M}_0$ in the expansion, we get

$$\tau_k^T\,\dot{m}_k + m_k = \chi_k^T\,h_k + \tau_k^T\,\gamma\,[\mathbf{M}\mathbf{H}]_k + (\partial M_k/\partial T)_H\vartheta, \tag{9}$$

where $\mathbf{h} = \mathbf{H} - \mathbf{H}_0$. The isothermal susceptibility χ_k^T is found from the derivatives

$$(\partial H_k/\partial M_k)_T = 1/\chi_k^T;\ (\partial H_k/\partial M_i)_T = 0 \text{ для } i \neq k, \tag{10}$$

taken at the equilibrium position. Here, $\chi_1^T = \chi_2^T = \chi_\perp^T$, $\chi_3^T = \chi_\parallel^T$, and the times of isothermal spin-spin relaxation τ_k^T are determined from

$$\tau_k^T = \chi_k^T/\lambda_k. \tag{11}$$

In the case of an isothermal process in the spin system, $\vartheta = 0$ and Eq. (9) describes the change in magnetization with time, without any additional conditions.

For adiabatic change of the state of the spin system, we find for the time of adiabatic spin-spin relaxation τ_k^S:

$$\tau_k^S = \chi_k^S/\lambda_k, \tag{12}$$

where χ_k^S is the adiabatic susceptibility. It is now easy to find that

$$\tau_k^S/\tau_k^T = \chi_k^S/\chi_k^T = C_M/C_H, \tag{13}$$

where C_H and C_M are the specific heats of the spin system, for constant H and M, respectively.

3. The quantity of heat dQ transferred to the spin system of the lattice in the time dt we set equal to

$$dQ = \alpha\vartheta dt.$$

Then, by Eq. (3),

$$\dot{\vartheta} + \frac{\vartheta}{\tau_M} = \frac{T_0}{C_M}\left(\frac{\partial \mathbf{H}}{\partial T}\right)_M \dot{\mathbf{m}}, \tag{14}$$

where $\tau_M = C_M/\alpha$ is the time of external, spin-lattice relaxation for constant magnetization. Considering the magnetization M in Eq. (14) as a function of H and T, we can put this equation in the form

$$\dot{\vartheta} + \frac{\vartheta}{\tau_H} = \frac{T_0}{C_H}\left(\frac{\partial \mathbf{M}}{\partial T}\right)_H \dot{\mathbf{h}}. \tag{15}$$

Here $\tau_H = C_H/\alpha$ is the time of external, spin-lattice relaxation for constant H.

Eliminating ϑ from Eqs. (9) and (14), after simple transformations, we get the equation for the change in the magnetization with time:

$$\tau_k^T \tau_M \ddot{m}_k + (\tau_H + \tau_k^T)\dot{m}_k + m_k = \tau_M \chi_k^T \dot{h}_k + \chi_k^T h_k + \tau_k^T \tau_M \gamma [\dot{\mathbf{M}}\mathbf{H}]_k + \tau_k^T \gamma [\mathbf{MH}]_k, \tag{16}$$

which takes into account the internal and the external relaxation.

In order to clarify the role of the spin-lattice relaxation, we rewrite the last equation in the form

$$\tau_H \frac{d}{dt}\{\tau_k^S \dot{m}_k + m_k - \chi_k^S h_k - \gamma\tau_k^S [\mathbf{MH}]_k\} + \{\tau_k^T \dot{m}_k + m_k - \chi_k^T h_k - \gamma\tau_k^T [\mathbf{MH}]_k\} = 0. \tag{17}$$

It follows from (13) that if we set $C_H = C_M$, then $\tau_k^S = \tau_k^T = \tau_k$, and therefore Eq. (17) can be written as

$$\tau_H dA/dt + A = 0,$$
$$A = \tau_k \dot{m}_k + m_k - \chi_k h_k - \gamma\tau_k [\mathbf{MH}]_k. \tag{18}$$

As is seen from (18), the spin-lattice relaxation in this case appears only in transient processes, since the stationary solution of (18)

$$\tau_k \dot{m}_k + m_k = \chi_k h_k + \gamma\tau_k [\mathbf{MH}]_k \tag{19}$$

takes only the spin-spin relaxation into account.

For ferromagnetics below the Curie point,[10] the ratio

$$\frac{C_H}{C_M} = 1 + H_0 \frac{\partial M}{\partial T} \Big/ \left(H_{ex}\frac{\partial M}{\partial T} - C_{dip}\right)$$

differs from unity only in the third decimal place, since the ratio of H_0 to the intensity of the field of exchange forces H_{ex} is of the order 10^{-3}, while the specific heat of the dipole interaction $C_{dip} \ll H_{ex}|\partial M/\partial T|$. Above the Curie point, the ratio

$$C_H/C_M = 1 + CH_0^2 T^{-2}/(CH_{ex}^2 T^{-2} + C_{dip}),$$

where C is the Curie constant, which differs from unity only in the sixth decimal place.

The closeness of the ratio of C_H/C_M to unity is brought about, as follows from what was said above, from the presence in the ferromagnetics of a strong field of exchange forces. Therefore, the specific heat of the spin system is so large that the radiofrequency field at small amplitude does not succeed in raising its temperature in any appreciable amount. As a result, the spin-lattice relaxation effect is shown to be insignificant and practically escapes observation.

On the other hand, in the case $\tau_H\omega_0 \gg 1$, where ω_0 is the resonance frequency, it is seen from (17) that, neglecting the group of terms in the second curly brackets, we obtain the equation

$$\tau_k^S \dot{m}_k + m_k = \chi_k^S h_k + \tau_k^S \gamma [\mathbf{MH}]_k,$$

which takes into account only the spin-spin relaxation. In similar fashion we obtain, in the other limiting case when $\tau_H\omega_0 \ll 1$:

$$\tau_k^T \dot{m}_k + m_k = \chi_k^T h_k + \tau_k^T \gamma [\mathbf{MH}]_k.$$

Therefore, for ferromagnetics, the spin-lattice relaxation can exist independently of the magnitude of the ratio C_H/C_M only upon satisfaction of the condition $\tau_H\omega_0 \approx 1$, which usually does not hold.

4. Neglecting the spin-lattice relaxation, we shall start out in what follows from Eq. (8) which, taking (2) into account, can be written in the form

$$\dot{M}_k = \lambda_k\left(H_k - \frac{H^*}{M} M_k\right) + \gamma\,[\mathbf{MH}]_k. \qquad (20)$$

Since $\mathbf{H}^*$ and $\mathbf{M}$ are connected by the equation of state, then, in the approximation assumed,

$$M/H^* = M_0/H_0 = \chi_0, \qquad (21)$$

where χ_0 is the static susceptibility and M_0 is the equilibrium magnetization corresponding to the field H_0. Introducing the longitudinal and transverse relaxation times

$$T_\perp = \chi_0/\lambda_\perp;\; T_\parallel = \chi_0/\lambda_\parallel, \qquad (22)$$

we rewrite (20), keeping (21) in mind:

$$\dot{M}_{x,y} = \gamma[\mathbf{MH}]_{x,y} - (M_{x,y} - \chi_0 H_{x,y})/T_\perp,$$
$$\dot{M}_z = \gamma\,[\mathbf{MH}]_z - (M_z - \chi_0 H_z)/T_\parallel. \qquad (23)$$

The equations obtained above differ from the Bloch equations (which are applicable in the theory of nuclear magnetic resonances) by the presence of the terms $\chi_0 H_x$ and $\chi_0 H_y$. In ferromagnetic substances, the vector $\mathbf{M}$ in weak radiofrequency fields and even for resonance is close in its direction to $\mathbf{M}_0$, as a consequence of the large width of the absorption lines. Therefore, the components M_x and M_y are small, and it is not possible to neglect the terms $\chi_0 H_x$ and $\chi_0 H_y$. The equations, in a form that coincides with (23), were set up in Ref. 11 for the description of the phenomenon of ferromagnetic resonance.

5. At temperatures far removed from the Curie point ($T < \Theta$), the external field H_i does not appreciably change the magnitude of the vector of spontaneous magnetization $\mathbf{M} = \mathbf{M}_s$, producing only a change in its direction. If we require constancy of the magnitude of the vector $\mathbf{M}$, then it follows from (8) that

$$\sum_{k=1}^{3} \lambda_k M_k (H_k - H_k^*) = 0 \qquad (24)$$

whence, by (2),

$$\mathbf{H}^* = \xi\,(\mathbf{MH})\,\mathbf{M}/M^2, \qquad (25)$$

where

$$\xi = \left[1 + \left(\frac{\lambda_\parallel}{\lambda_\perp} - 1\right)\frac{M_z H_z}{\mathbf{MH}}\right] \Big/ \left[1 + \left(\frac{\lambda_\parallel}{\lambda_\perp} - 1\right)\left(\frac{M_z}{M}\right)^2\right]. \qquad (26)$$

In this case, we can write Eq. (8) in the form

$$\dot{M}_{x,y} = \gamma\,[\mathbf{MH}]_{x,y} - \lambda_\perp M^{-2}\,[\mathbf{M}\,[\mathbf{MH}]]_{x,y}$$
$$+ \lambda_\perp (1 - \xi)\,(\mathbf{MH})\,M^{-2} M_{x,y};$$
$$\dot{M}_z = \gamma\,[\mathbf{MH}]_z - \lambda_\parallel M^{-2}\,[\mathbf{M}\,[\mathbf{MH}]]_z \qquad (27)$$
$$+ \lambda_\parallel (1 - \xi)\,(\mathbf{MH})\,M^{-2} M_z.$$

For $\lambda_\perp = \lambda_\parallel = \lambda$, Eqs. (28) transform to the Landau-Lifshitz equations, as is seen from Eq. (26). The Landau-Lifshitz equations are widely used in the theory of ferromagnetic resonance. On the other hand, in weak radiofrequency fields, where $h \ll H_0$,

$$M_z H_z/\mathbf{MH} \approx 1; \quad M_z^2/M^2 \approx 1$$

the coefficient $\xi \approx 1$, and the equations (27) are simplified:

$$\dot{M}_{x,y} = -\lambda_\perp M^{-2}\,[\mathbf{M}\,[\mathbf{MH}]]_{x,y} + \gamma\,[\mathbf{MH}]_{x,y},$$
$$\dot{M}_z = -\lambda_\parallel M^{-2}\,[\mathbf{M}\,[\mathbf{MH}]]_z + \gamma\,[\mathbf{MH}]_z. \qquad (28)$$

The right side of the equation for $\dot{M}_z$ is equal to zero in this case; the solutions of Eq. (28) do not contain the constant λ, and coincide with the solutions of the Landau-Lifshitz equation in the case of weak fields for $\lambda_\perp = \lambda$.

If we assume that $\lambda_\perp \neq \lambda_\parallel$, then the difference of (23) from the Landau-Lifshitz equation can exist only in the case of strong radiofrequency fields, where $\xi \neq 1$.

Since the solution of the Landau-Lifshitz equation and Eq. (23) coincide in the case of weak rf fields, and they also coincide with Eq. (27), then the phenomenon of ferromagnetic resonance in weak fields is shown to be very insensitive to the detailed form of the equation employed for their description. Preference for this or that form of the equation can be made only upon observation of nonlinear effects, for example, observation of the change of the z component of the magnetization.

[1] L. D. Landau and E. M. Lifshitz, Phys. Z. Sowjetunion **8**, 153 (1935).

[2] F. Bloch, Phys. Rev. **70**, 460 (1946).

[3] R. Wangsness, Phys. Rev. **104**, 857 (1956).

[4] C. J. Gorter, Paramagnetic Relaxation (Elsevier, New York, 1947).

[5] I. G. Shaposhnikov, J. Exptl. Theoret. Phys. (U.S.S.R.) **18**, 533 (1948).

[6] I. G. Shaposhnikov, J. Exptl. Theoret. Phys. (U.S.S.R.) **19**, 225 (1949).

[7] I. G. Shaposhnikov, Uc. Zap. Molotov State Univ., vol. 8, No. 1 (1953).

[8] M. A. Leontovich, Введение в термодинамику (Introduction to Thermodynamics), GITTL, 1956.

[9] S. R. deGroot, Thermodynamics of Irreversible Processes (Interscience, New York, 1951).

[10] N. Bloembergen and S. Wang, Phys. Rev. **93**, 72 (1954).

[11] R. Wangsness and F. Bloch, Phys. Rev. **89**, 728 (1953).

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139