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Abstract

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PHYSICS

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ON THE KINETICS OF MAGNETIZATION OF A PARAMAGNET

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1. Let us consider the problem: to find the time dependence of the macroscopic magnetic moment $\mathbf{M}(t)$ of a paramagnet under the action of an applied magnetic field $\mathbf{H}(t)$, whose time dependence is given.
2. We shall use the scheme proposed by Robertson ⁽¹⁾ for deriving equations of motion for macroscopic physical quantities (this scheme had already been used by its author ⁽²⁾ to consider certain questions of magnetic kinetics). We restrict ourselves to the case in which, in the large physical system under consideration, there is no macroscopic mechanical motion and all macroscopic quantities characterizing it are homogeneous in space. Let $F_1(t), F_2(t), \dots$ denote any of these quantities, mutually independent and, together with the external parameters $V_1(t), V_2(t), \dots$, providing a complete macroscopic description of the states traversed by the system under consideration in the process of interest to us. Let $F_r = \bar{f}_r$, $r = 1, 2, \dots$, where f_r are certain microscopic quantities whose operators $\hat{f}_r$ do not contain time, and the bar denotes averaging over the mentioned states. The functions $V_r(t)$, $r = 1, 2, \dots$, are given; the functions $F_r(t)$, $r = 1, 2, \dots$, are to be determined. Scheme ⁽¹⁾ leads to the following exact equations of motion for $F_r(t)$, $r = 1, 2, \dots$:

$$\dot{F}_r(t) = -i \text{Sp} [\hat{f}_r \hat{L}(t) \hat{\sigma}(t)] - \int_{t_0}^t dt' \text{Sp} \{ \hat{f}_r \hat{L}(t) \hat{T}(t, t') [\hat{1} - \hat{P}(t')] \hat{L}(t') \hat{\sigma}(t') \} \quad (1)$$

under the condition that

$$\hat{\rho}(t_0) = \hat{\sigma}(t_0). \quad (2)$$

Here $\hat{L}(t) \dots \equiv [\hat{\mathcal{H}}(t), \dots]$, where $\hat{\mathcal{H}}(t)$ is the Hamiltonian of the system and $[\ , \]$ is the commutator; $\hat{\rho}$ is the statistical operator of the system;

$$\hat{\sigma}(t) \equiv \{\text{Sp} \exp[-\Sigma \beta_r(t) \hat{f}_r]\}^{-1} \exp[-\Sigma \beta_r(t) \hat{f}_r], \quad (3)$$

where $\beta_r(t) = \beta_r[F_1(t), F_2(t), \dots]$, $r = 1, 2, \dots$, are macroscopic quantities determined by the system of equations

$$\text{Sp}(\hat{\sigma} \hat{f}_r) = F_r, \quad r = 1, 2, \dots; \quad (4)$$

$\hat{1}$ is the unit operator;

$$\hat{P}(t) \dots \equiv \Sigma[\partial \hat{\sigma}(t)/\partial F_r(t)] \text{Sp}(\hat{f}_r \dots); \quad (5)$$

$\hat{T}(t, t')$ is determined by the equation

$$\frac{\partial}{\partial t'} \hat{T}(t, t') = i \hat{T}(t, t') [\hat{1} - \hat{P}(t')] \hat{L}(t') \quad (6)$$

with the initial condition $\hat{T}(t, t) = \hat{1}$. Equations (1) are obtained directly from the Neumann equation $\dot{\hat{\rho}} = -i \hat{L} \hat{\rho}$ (we have set $\hbar = 1$). The operator $\hat{\sigma}$ is not the statistical operator of the system and, consequently, does not satisfy

the Neumann equation; it enters into the consideration when determining the quantities β_r (we shall call them conjugate to the quantities F_r) by equations (4), which thus contain no physical assumptions.

3. It is of interest to study processes that begin from equilibrium, so that $\hat{\rho}(t_0) = \{\text{Sp} \exp[-T^{-1} \hat{\mathcal{H}}(t_0)]\}^{-1} \exp[-T^{-1} \hat{\mathcal{H}}(t_0)]$, where T is the temperature (we set $k = 1$). In order that scheme (1) be suitable for considering such processes, it must be that $\sum \beta_r(t_0) \hat{f}_r = T^{-1} \hat{\mathcal{H}}(t_0)$. We shall make, however, more special but physically clearer assumptions: we shall assume that

$$\hat{\mathcal{H}}(t) = \sum a_r(t) \hat{f}_r \equiv \sum \hat{\mathcal{H}}_r(t), \quad (7)$$

where the quantities $a_r(t)$ characterize the external conditions in which the system under consideration is found (and, consequently, are expressed in terms of the external parameters $V_r(t)$), and that in equilibrium

$$\beta_r^{\text{eq}} = a_r^{\text{eq}} T^{-1}, \quad (8)$$

so that $\hat{\sigma}^{\text{eq}} = \hat{\rho}^{\text{eq}}$, by which, obviously, (2) is ensured for processes of the type considered. We shall use the following terminology: of the system with $\hat{\mathcal{H}}$ from (7) we shall say that it consists of subsystems (parts) with Hamiltonians $\hat{\mathcal{H}}_r$, and the quantities β_r^{-1} , inversely conjugate to $F_r = \hat{f}_r$, we shall call the generalized

temperatures of these subsystems; if for any subsystems $a_r = 1$, i.e. $\hat{\mathcal{H}}_r = \hat{f}_r$, then the corresponding quantities β_r^{-1} will be called the temperatures of these subsystems.

4. Let us now turn to the problem posed in Sec. 1 within the restrictions adopted at the beginning of Sec. 2. We shall assume that the only external action on the paramagnetic sample is the action of the applied field, and we shall write the Hamiltonian of the sample in this field in the form

$$\hat{\mathcal{H}}(t) = -\mathbf{H}(t) \cdot \hat{\mathcal{M}} + \hat{\mathcal{U}} + \hat{\mathcal{H}}_L, \quad (9)$$

where $\hat{\mathcal{M}}$ is the operator of the total magnetic moment of the paramagnet; all nonmagnetic degrees of freedom are assigned to $\hat{\mathcal{H}}_L$, while $\hat{\mathcal{U}}$ includes all internal interactions connected with the magnetic degrees of freedom. The first of the subsystems singled out in this way we shall call the Zeeman subsystem, the second the interaction subsystem, and the third the lattice; the aggregate of the Zeeman subsystem and the interaction subsystem may be called the spin system. Up to the initial instant t_0 the applied field did not change, and the paramagnet was in equilibrium; beginning from t_0 , the field changes with time in a prescribed way. We are interested in $\mathbf{M}(t)$ for $t > t_0$.

It is convenient to write the sum $\sum \beta_r \hat{f}_r$ entering into (3) in the form

$$\sum \beta_r \hat{f}_r = \beta_M \cdot \hat{\mathcal{M}} + \beta \hat{\mathcal{U}} + \beta_L \hat{\mathcal{H}}_L \equiv \beta \left(-\mathbf{H}^* \cdot \hat{\mathcal{M}} + \hat{\mathcal{U}} + \hat{\mathcal{H}}_L^* \right), \quad (10)$$

where

$$\mathbf{H}^* \equiv -\beta^{-1} \beta_M, \quad \hat{\mathcal{H}}_L^* \equiv \beta^{-1} \beta_L \hat{\mathcal{H}}_L; \quad (11)$$

according to (8), in equilibrium $\beta_M^{\text{eq}} = -T^{-1} \mathbf{H}$, $\beta^{\text{eq}} = \beta_L^{\text{eq}} = T^{-1}$, $\mathbf{H}^{*\text{eq}} = \mathbf{H}$, $\hat{\mathcal{H}}_L^{*\text{eq}} = \hat{\mathcal{H}}_L$. Bearing in mind the possibility of the presence of natural anisotropy in the paramagnet, we put

$$\hat{\mathcal{M}} = \bar{g} \cdot \mathbf{I}, \quad (12)$$

where $\mathbf{I}$ is the total mechanical angular momentum and $\bar{g}$ is the gyromagnetic tensor.

5. Now one could write (1) for $\mathbf{M} = \hat{\mathcal{M}}$, $U \equiv \hat{\mathcal{U}}$, $H_L \equiv \hat{\mathcal{H}}_L$ as the F_r . However, as applied to our problem it is more expedient to use the equations of motion for $F_r = \mathbf{M}, U, H_L$, somewhat dif-

differing from (1), which are obtained if one repeats the consideration leading in scheme (1) to equations (1), but instead of the operator $\hat{P}$ from (5) one uses the operator

$$\hat{\mathcal{P}} \dots \equiv (\partial \hat{\sigma} / \partial \mathbf{M}) \cdot \text{Sp}(\hat{\mathcal{M}} \dots) + (\partial \hat{\sigma} / \partial H_L) \text{Sp}(\hat{\mathcal{H}}_L \dots), \quad (13)$$

which differs from $\hat{P}$ by the absence of the term with $\hat{U}$. With the aid of (12) and of the relation

$$\dot{U} = \mathbf{H} \cdot \dot{\mathbf{M}} - \dot{H}_L, \quad (14)$$

which follows directly from (9) and Neumann's equation, scheme (1), modified in the indicated way, gives

$$\begin{aligned} \dot{F}(t) = \delta_{F\mathbf{M}} |\bar{g}| \left\{ \left[\left(\bar{g}^{-1} \cdot \bar{\sigma}^{-1} \right) \mathbf{M} \right] \times [\mathbf{H}(t) - \mathbf{H}^*] \right\} \\ - \int_{-\infty}^{\infty} dt' K_{\hat{f}\hat{\mathcal{M}}}(t, t') [\mathbf{H}^*(t') - \mathbf{H}(t')] \\ - \int_{-\infty}^{\infty} dt' K_{\hat{f}\hat{\mathcal{H}}_L}(t, t') [\beta(t') - \beta_L(t')] \end{aligned} \quad (15)$$

with $F \equiv \mathbf{M}, H_L$ and $\hat{f} \equiv \hat{\mathcal{M}}, \hat{\mathcal{H}}_L$; here

$$\begin{aligned} K_{\hat{f}\hat{\mathcal{M}}}(t, t') \equiv \theta(t - t') \text{Sp}\{\hat{f}\hat{L}(t)\hat{T}(t, t')[\hat{1} - \hat{\mathcal{P}}(t')][\hat{\mathcal{M}}, \hat{\sigma}(t')]\} \\ + i \int_{-\infty}^{\infty} d\tau d\tau' \theta(t - \tau) \text{Sp}\{\hat{f}\hat{L}(\tau)\hat{T}(\tau, \tau')[\partial \hat{\sigma}(\tau) / \partial U(\tau)]\} u(\tau, \tau') \times \\ \times \left\{ -\theta(\tau' - t') |\bar{g}| \left[\left(\bar{g}^{-1} \cdot \bar{\sigma}^{-1} \right) \cdot \mathbf{M}(t') \right] \times \mathbf{H}(t') \right\} \\ + \theta(\tau' - t') \text{Sp}(\hat{U}\hat{L}(\tau')\hat{T}(\tau', t')[\hat{1} - \hat{\mathcal{P}}(t')][\hat{\mathcal{M}}, \hat{\sigma}(t')]) \}, \end{aligned} \quad (16)$$

$$\begin{aligned} K_{\hat{f}\hat{\mathcal{H}}_L}(t, t') \equiv \{\theta(t - t') \text{Sp}(\hat{f}\hat{L}(t)\hat{T}(t, t')[\hat{1} - \hat{\mathcal{P}}(t')][\hat{\mathcal{H}}_L, \hat{\sigma}(t')]) \\ + i \int_{-\infty}^{\infty} d\tau d\tau' \theta(t - \tau) \theta(\tau' - t') \text{Sp}\{\hat{f}\hat{L}(\tau)\hat{T}(\tau, \tau')[\partial \hat{\sigma}(\tau) / \partial U(\tau)]\} u(\tau, \tau') \times \\ \times \text{Sp}(\hat{U}\hat{L}(\tau')\hat{T}(\tau', t')[\hat{1} - \hat{\mathcal{P}}(t')][\hat{\mathcal{H}}_L, \hat{\sigma}(t')])\} \beta^{-1}(t'). \end{aligned} \quad (17)$$

$\theta(t - t') = 1$ for $t' < t$ and 0 for $t' > t$; the function $u(t, t')$ is determined by the equation

$$\int_{-\infty}^{\infty} d\tau u(t, \tau) u^{-1}(\tau, t') = \delta(t - t'), \quad (18)$$

where

$$u^{-1}(t, t') = \delta(t - t') - i\theta(t - t') \text{Sp}\{\hat{U}\hat{L}(t)\hat{T}(t, t')[\partial\hat{\sigma}(t')/\partial U(t')]\}, \quad (19)$$

and, for the convenience of subsequent calculations, $t_0 = -\infty$ has been adopted. The integro-differential equations (15), in which $\mathbf{H}^*$, β , and β_L must be expressed in terms of $\mathbf{M}$, U , H_L according to the general scheme (1), together with relation (14), give the solution of the problem under discussion.

6. Equations (15) are exact; in particular, in their derivation it was not assumed that the states passed through by the system are, in any sense, close to equilibrium. However, in order to apply these equations to concrete cases it is necessary to calculate the quantities K , and this, of course, is feasible only in one or another approximation. Let us list here some circumstances that may give rise to different approximations: if $|\beta_M|$ and β are small, then in the expansion of $\hat{\sigma}$ in β one may restrict oneself to one or two steps (the high-temperature approximation); if the lattice may be regarded as remaining in equilibrium all the time, then $\beta_L = \text{const}(t)$, which substantially simpli-

poses the problem; if $\mathbf{H}(t)$ varies slowly, memory effects may be neglected, and the equations of the problem become differential; if the variable part $\mathbf{H}(t)$ is small, the contribution it causes to the time dependence of the quantities K becomes negligible. Of course, in each concrete case the possibility of one or another of these approximations must be justified by an appropriate quantitative estimate.

7. It can be shown that, if only the approximations just enumerated are possible, then (15) and (14) lead to the following equations for finding $\mathbf{M}(t)$:

$$\dot{\mathbf{M}} = g [(\bar{c}^{-1} \cdot \mathbf{M}) \times (\mathbf{H} - \bar{N} \cdot \mathbf{M})] - \bar{\kappa} \cdot (\mathbf{M} - \beta \bar{c} \cdot \mathbf{H}) - \chi(\beta - \beta_0); \quad (20)$$

$$\dot{\beta} = b^{-1} \mathbf{H} \cdot \dot{\mathbf{M}} = -\alpha(\beta - \beta_0) - \lambda \cdot (\mathbf{M} - \beta \bar{c} \cdot \mathbf{H}); \quad (21)$$

here

$$g = \frac{1}{3} n j(j+1) |g|, \quad (22)$$

where n is the number of magnetic particles, assumed identical and in equivalent positions, and j is the quantum number of the angular momentum of an individual particle; $\bar{c}$ and $\bar{N}$ are characteristics of the paramagnet entering into the equilibrium magnetic susceptibility:

$$\bar{\chi}^{\text{eq}} = \beta \bar{c} - \beta^2 \bar{c} \cdot \bar{N} \cdot \bar{c} + \dots, \quad (23)$$

where the dots denote further terms of the expansion in β ; b is the constant of magnetic heat capacity, which enters into the expression for V :

$$U = -b\beta + \dots; \quad (24)$$

β_0 denotes the constant value of β_L ; $\bar{\kappa}$, χ , α , λ are kinetic coefficients (there is a relation between χ and λ following from the Onsager relations). In deriving equations (20) and (21), the second order of the expansion in β has been taken into account only in the dynamical term of equation (20), where neglect of the term with $\bar{N}$ would lead to the exclusion from consideration of new effects due to the fact that the direction of the second factor in the vector product does not coincide with the direction of $\mathbf{H}$. For a paramagnet without natural anisotropy ($\bar{c} = c\mathbf{1}$, $\bar{N} = N\mathbf{1}$), equations (20) and (21) coincide with the corresponding equations of the most general variant of the phenomenological theory of paramagnetic relaxation (see ⁽³⁾), which, in turn, contain as special cases the Bloch equation and all its known modifications. It should be noted, however, that the equations of the phenomenological theory are obtained for small values characterizing the nonequilibrium of the quantities $\mathbf{M} - \beta c\mathbf{H}$ and $\beta - \beta_0$, whereas equations (20) and (21) do not assume such a restriction.

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