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MAGNETIZATION OF A FERRODIELECTRIC ALONG AN AXIS OF DIFFICULT MAGNETIZATION

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It is shown that the contradictions that arise in calculation of the magnetization of a ferrodielectric in the direction of an axis of difficult magnetization at low temperatures are due to an approximation made in the calculation of the free energy. A somewhat different method of calculating the magnetization is proposed; the magnetization, it turns out, is a continuous function of the field both in the case of a positive and in the case of a negative anisotropy constant. The magnetic susceptibility has a logarithmic singularity at a field equal to the anisotropy field in the case of a positive anisotropy constant, and a finite discontinuity in the case of a negative anisotropy constant.

CALCULATION of the magnetization of a ferrodielectric at low temperatures along a difficult direction, by means of the thermodynamic relation $M = -\partial\mathfrak{F}/\partial H$ ($\mathfrak{F}$ is the free energy per unit volume, H is the constant magnetic field) leads to a contradictory result, if $\mathfrak{F}$ is determined in the spin-wave approximation. The contradiction is connected with the fact that if the field H increases, remaining less than the anisotropy field H_A, then at a nonvanishing temperature the magnetization becomes larger than the nominal value. Furthermore, the values of the magnetization at fields larger and smaller than H_A do not agree in the limit $H \to H_A$ (see, for example,[1]).

In the present paper, a somewhat different method of calculating the magnetization is proposed; with this method, the contradictions mentioned do not arise.

The point is that in finding the magnetization along a difficult direction, because of the impossibility of an exact solution of the problem, a semiclassical treatment is used, in which the ground state is characterized by a field-dependent direction of equilibrium of the magnetization with respect to the easy axis. The behavior of the system near the ground state is considered in the approximation of small oscillations of the vector magnetization about the equilibrium position. In this connection there arises a necessity for a transformation to a new system of coordinates, one of whose axes coincides with the equilibrium direction of the moment. As a result, there occurs a dependence of the Hamiltonian on the field both explicitly and through the angle of rotation. If rotation about the x axis occurs, then the Hamiltonian $\hat{\mathscr{H}}'$ in the new system of coordinates is connected with the original Hamiltonian $\hat{\mathscr{H}}$ by the relation

$$\hat{\mathscr{H}} = e^{-i\hat{s}_x\theta}\hat{\mathscr{H}}'e^{i\hat{s}_x\theta},$$

where s_x is the component of the total spin along the x axis and θ is the angle of rotation. On using for the free energy the expression $\mathfrak{F} = -T\ln \operatorname{Sp} e^{-\mathscr{H}/T}$, we get

$$M = -\operatorname{Sp}\frac{\partial\hat{\mathscr{H}}}{\partial H}e^{-\hat{\mathscr{H}}/T}/\operatorname{Sp}e^{-\hat{\mathscr{H}}/T}.$$

On taking account of the relation between $\hat{\mathscr{H}}$ and $\hat{\mathscr{H}}'$, we find

$$\frac{\partial\hat{\mathscr{H}}}{\partial H} = i\frac{\partial\theta}{\partial H}[\hat{\mathscr{H}}\hat{s}_x] + e^{-i\hat{s}_x\theta}\frac{\partial\hat{\mathscr{H}}'}{\partial H}e^{i\hat{s}_x\theta}.$$

Averaging of the first term, with a Gibbs distribution, gives zero. Therefore

$$M = -\operatorname{Sp}e^{-i\hat{s}_x\theta}\frac{\partial\hat{\mathscr{H}}'}{\partial H}e^{i\hat{s}_x\theta}e^{-\hat{\mathscr{H}}/T}/\operatorname{Sp}e^{-\hat{\mathscr{H}}/T}. \quad (1)$$

In other words, in an exact treatment a transformation to a new system of coordinates does not lead to any additional terms in the magnetization; this is as it should be. If, however, the free energy is calculated in the spin-wave approximation, then as a result of the rotation there appear additional terms, which lead to the contradictions mentioned.

The calculations made below, which make use of formula (1) in the spin-wave approximation also, lead to results that show that the magnetization is a continuous function of the field both for positive and for negative anisotropy; the magnetic susceptibility, however, has in the case of a positive anisotropy constant a logarithmic singularity at $H = H_A$, and in the negative case a finite discontinuity.

1. THE CASE OF A POSITIVE ANISOTROPY CONSTANT ($k > 0$)

1. We choose the axis of easiest magnetization as the z axis of the system of coordinates; as y axis, we take the direction of the constant magnetic field **H**, which coincides with a direction of difficult magnetization.

We write the Hamiltonian of the system in the form[2]

$$\hat{\mathscr{H}} = -{}^1/_2\sum_{l,m}\hat{s}_l\hat{s}_m J_{lm} + K\mu^2\sum_l[(\hat{s}_l^x)^2 + (\hat{s}_l^y)^2] - \mu H\sum_l\hat{s}_l^y. \quad (2)$$

Here $\hat{\mathbf{s}}_l$ is the spin operator at the l-th site, and μ is the Bohr magneton.

The first term in (2) describes the exchange interaction (J_{lm} is the exchange integral). The second term is the magnetic-anisotropy energy ($K \sim \beta/a^3$, where β is the dimensionless anisotropy constant and a is the lattice constant). The third term is the energy of interaction of the spin system with the constant magnetic field. For simplicity, we take no account of dipole-dipole interaction. (The results obtained below are still valid qualitatively when account is taken of dipole-dipole interaction.)

By use of the relation

$$\hat{s}_l^2 = (\hat{s}_l^x)^2 + (\hat{s}_l^y)^2 + (\hat{s}_l^z)^2$$

the Hamiltonian can be written thus:

$$\hat{\mathcal{H}} = -{}^1/_2 \sum_{l,m} J_{lm}\hat{s}_l\hat{s}_m - K\mu^2 \sum_l (\hat{s}_l^z)^2 - \mu H \sum_l \hat{s}_l^y. \quad (3)$$

For determination of the ground state of the Hamiltonian (3), we shall consider the spins to be c-numbers; this is justified for sufficiently large spins. Then the equilibrium value s_0 of a spin s makes some angle θ with the axis of easiest magnetization (s_0 lies in the zy plane). This angle is determined by minimization of the energy

$$E_0 = -{}^1/_2 \sum_{l,m} J_{lm}s^2 - K\mu^2 s^2 N \cos^2\theta - \mu H s N \sin\theta, \quad (4)$$

which corresponds to a uniform magnetization $s_l \equiv s$ (N is the total number of spins in the system).

As a result we get

$$\sin\theta = H/H_A \text{ for } H \leqslant H_A, \quad (5)$$

$$\cos\theta = 0 \text{ for } H \geqslant H_A. \quad (6)$$

Here the anisotropy field $H_A \equiv 2K\mu s$.

2. We consider the range of fields $H \le H_A$. We introduce a primed system of coordinates, such that the z' axis is directed along s_0 and the y' axis lies in the zy plane. Then the connection between the spin-projection operators in these systems will be given by the relations

$$\begin{aligned} \hat{s}_l^x &= \hat{s}_l^{x'}, \\ \hat{s}_l^y &= \hat{s}_l^{z'}\sin\theta + \hat{s}_l^{y'}\cos\theta, \\ \hat{s}_l^z &= \hat{s}_l^{z'}\cos\theta - \hat{s}_l^{y'}\sin\theta. \end{aligned} \quad (7)$$

Following Holstein and Primakoff [3], we introduce the Bose operators $\hat{a}_l$ and $\hat{a}_l^+$, which satisfy the commutation rules

$$\begin{aligned} \hat{a}_l\hat{a}_m^+ - \hat{a}_m^+\hat{a}_l &= \delta_{ml}, \\ [\hat{a}_l\hat{a}_m] = [\hat{a}_l^+,\ \hat{a}_m^+] &= 0, \end{aligned} \quad (8)$$

and we express the operators $\hat{s}_{x'}$, $\hat{s}_{y'}$, and $\hat{s}_{z'}$ in terms of them, retaining only terms linear and quadratic in the operators $\hat{a}_l$ and $\hat{a}_l^+$:

$$\begin{aligned} \hat{s}_l^{x'} &= \sqrt{s/2}\,(\hat{a}_l^+ + \hat{a}_l), \\ \hat{s}_l^{y'} &= i\sqrt{s/2}\,(\hat{a}_l^+ - \hat{a}_l), \\ \hat{s}_l^{z'} &= s - \hat{a}_l^+\hat{a}_l. \end{aligned} \quad (9)$$

Then the operators $\hat{s}_l^x$, $\hat{s}_l^y$, and $\hat{s}_l^z$ take the following form:

$$\begin{aligned} \hat{s}_l^x &= \sqrt{s/2}\,(\hat{a}_l^+ + \hat{a}_l), \\ \hat{s}_l^y &= (s - \hat{a}_l^+\hat{a}_l)\sin\theta + i\sqrt{s/2}\,(\hat{a}_l^+ - \hat{a}_l)\cos\theta, \\ \hat{s}_l^z &= (s - \hat{a}_l^+\hat{a}_l)\cos\theta - i\sqrt{s/2}\,(\hat{a}_l^+ - \hat{a}_l)\sin\theta. \end{aligned} \quad (10)$$

After substitution of (10) in (2) and use of the commutation rules (8), with subsequent introduction of the Fourier components of the operators $\hat{a}_l^+$ and $\hat{a}_l$, we get $\hat{\mathcal{H}}$ in the form (the terms linear in the Bose operators drop out because of the choice of the ground state)[1]

$$\hat{\mathcal{H}} = E_0 - \frac{\mu H_A}{4} N\left(\frac{H}{H_A}\right)^2 + \sum_{\mathbf{k}} (A_{\mathbf{k}}\hat{a}_{\mathbf{k}}^+\hat{a}_{\mathbf{k}}^+ + {}^1/_2 B\hat{a}_{\mathbf{k}}^+\hat{a}_{-\mathbf{k}}^+ + {}^1/_2 B\hat{a}_{\mathbf{k}}\hat{a}_{-\mathbf{k}}),$$

where

$$A_{\mathbf{k}} = \frac{\mu H_A}{2}\left[2 - \left(\frac{H}{H_A}\right)^2\right] + Js(ka)^2,$$

$$B = \frac{\mu H_A}{2}\left(\frac{H}{H_A}\right)^2. \quad (12)$$

We now diagonalize the Hamiltonian by means of the u, v representation [4]:

$$\hat{a}_{\mathbf{k}} = u_{\mathbf{k}}\hat{c}_{\mathbf{k}} + v_{\mathbf{k}}\hat{c}_{-\mathbf{k}}^+, \qquad \hat{a}_{\mathbf{k}}^+ = u_{\mathbf{k}}\hat{c}_{\mathbf{k}}^+ + v_{\mathbf{k}}\hat{c}_{-\mathbf{k}}. \quad (13)$$

We get as a result

$$\mathcal{H} = \mathcal{H}_0 + \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}}\hat{c}_{\mathbf{k}}^+\hat{c}_{\mathbf{k}}, \quad (14)$$

where $\mathcal{H}_0$ is the energy of the ground state, with allowance for zero-point oscillations, and where

$$\varepsilon_{\mathbf{k}} = \sqrt{A_{\mathbf{k}}^2 - B^2}. \quad (15)$$

The coefficients of the u, v representation have the following form:

$$\begin{aligned} u_{\mathbf{k}} &= \sqrt{(A_{\mathbf{k}} + \varepsilon_{\mathbf{k}})/2\varepsilon_{\mathbf{k}}}, \\ v_{\mathbf{k}} &= -\sqrt{(A_{\mathbf{k}} - \varepsilon_{\mathbf{k}})/2\varepsilon_{\mathbf{k}}}. \end{aligned} \quad (16)$$

The mean value of the magnetization M in the direction of the axis of difficult magnetization is determined by formula (1).

On using (10), (15), and (16), we get the following result:

$$M_y(T) = M_0\frac{H}{H_A} - \frac{H}{H_A}\frac{\mu}{4\pi^2}\int_0^\infty \frac{A_{\mathbf{k}} - \varepsilon_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}} k^2\,dk - \frac{H}{H_A}\frac{\mu}{2\pi^2}\int_0^\infty \frac{A_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}}\frac{k^2\,dk}{e^{\varepsilon_{\mathbf{k}}/T} - 1}, \quad (17)$$

where

$$M_y(T) = \frac{\mu}{V}\left\langle \sum_l s_l^y \right\rangle, \qquad M_0 = \frac{N\mu s}{V}.$$

At zero temperature, the last term in formula (17) drops out, and as a result we get

$$M_y(0) = M_0\frac{H}{H_A}\left(1 - \frac{\mu}{4\pi^2 M_0}\int_0^\infty \frac{A_{\mathbf{k}} - \varepsilon_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}} k^2\,dk\right), \quad (18)$$

whence the magnetic susceptibility χ at $T = 0$ is

$$\chi(0) = \frac{M_0}{H_A} - \frac{\mu}{4\pi^2 H_A}\int_0^\infty \frac{A_{\mathbf{k}} - \varepsilon_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}} k^2\,dk - \left(\frac{H}{H_A}\right)^2 \frac{\mu^2}{4\pi^2}\int_0^\infty \frac{(A_{\mathbf{k}} + B)B}{\varepsilon_{\mathbf{k}}^2} k^2\,dk. \quad (19)$$

For $H \ll H_A$, the susceptibility $\chi(0) = M_0/H_A$; that is, at zero field the value of χ coincides with the value obtained from a classical treatment. With increase of H, the change of χ with field, as is evident from (19), is more complicated than the classical.

At $H = H_A$, the susceptibility has a logarithmic singularity:

$$\chi = \chi_{\text{reg}} + \frac{1}{16\pi^2}\sqrt{\frac{\mu H_A}{Js}}\frac{\mu M_0}{Js}\ln\left(1 - \frac{H}{H_A}\right), \quad (20)$$

where χ_{reg} is the part of the susceptibility that is finite at $H = H_A$.

3. We consider the range of fields $H \ge H_A$. The equilibrium value of θ is now determined by the condition (6), which gives $\theta = \pi/2$.

We again introduce a primed system of coordinates, with the z' axis directed along s_0 (s_0 in the unprimed

[1] We remark that in [1] the minimization with respect to the angle θ is carried out on a thermodynamic potential Ω (at a nonvanishing temperature) calculated by means of the Hamiltonian (2.1), in which linear terms are absent but the angle θ has not yet been determined. Since the terms linear in the operators $\hat{a}$ and $\hat{a}^+$ drop out of the Hamiltonian only for a quite definite θ, such a procedure seems to us incorrect.

system of coordinates is directed along the y axis, over the whole range of fields now under consideration). The relation between quantities in the primed and the unprimed systems will be the following:

$$\hat{s}_x = \hat{s}_{x'}, \qquad \hat{s}_y = \hat{s}_{z'}, \qquad \hat{s}_z = -\hat{s}_{y'}. \tag{21}$$

By a procedure analogous to the case $H \le H_A$, we get

$$\mathcal{H} = \mathcal{H}_0 + \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} \hat{c}_{\mathbf{k}}^{+} \hat{c}_{\mathbf{k}}, \tag{22}$$

where $\epsilon_{\mathbf{k}}$ is determined by formula (15), in which $A_{\mathbf{k}}$ and B have the following form:

$$A_{\mathbf{k}} = Js(ka)^2 - \frac{\mu H_A}{2}\left(1 - 2\frac{H}{H_A}\right), \qquad B = \mu H_A / 2. \tag{23}$$

The quantities $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ are given, as before, by the expressions (16).

With the aid of formula (1) we get the following value for the magnetic-moment density:

$$M_y = M_0 - \frac{\mu}{4\pi^2}\int_0^\infty \frac{A_{\mathbf{k}} - \varepsilon_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}} k^2\,dk - \frac{\mu}{2\pi^2}\int_0^\infty \frac{A_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}} \frac{k^2\,dk}{e^{\varepsilon_{\mathbf{k}}/T} - 1}. \tag{24}$$

From a comparison of (23) and (24) with (12) and (17), respectively, it is clear that the limiting values of the magnetization at $H = H_A$, as H approaches H_A from below and from above, agree. This means that the magnetization is continuous at $H = H_A$. Since the magnetic susceptibility, as is clear from (20), has a logarithmic singularity at $H = H_A$, this value of the field can be regarded as a point of phase transition of the second kind with respect to the field. It is easy to show that when H approaches H_A from above, χ also diverges logarithmically:

$$\chi = -\frac{1}{32\pi^2}\frac{\mu M_0}{Js}\sqrt{\frac{\mu H_A}{Js}}\ln\left(\frac{H}{H_A} - 1\right). \tag{25}$$

From a comparison of (25) with (20) it is clear, first, that the singular parts of χ differ by a factor 2; and, second, that the regular part is absent in (25).

When the field H is large in comparison with H_A, the magnetization at zero temperature approaches M_0, as it should:

$$M_0 - M = \frac{\mu}{128\pi a^3}\left(\frac{\mu H_A}{Js}\right)^2\sqrt{\frac{Js}{\mu H}}. \tag{26}$$

The temperature part of the magnetization in both magnetic-field regions gives a small negative contribution, according to the temperature, so that for arbitrary temperature $T \ll Js$ and at arbitrary fields, the value of the magnetization does not exceed M_0.

2. THE CASE OF NEGATIVE ANISOTROPY CONSTANT

If the anisotropy constant is negative, then the equilibrium direction of the moment in the absence of a field is perpendicular to the distinguished axis of the ferromagnet. We choose the equilibrium direction of the magnetic moment as the y axis and the direction of the magnetic moment as the z axis and the direction of the constant magnetic field H as the z axis (H will thereby be directed along a difficult axis). The Hamiltonian of our system in this case is written thus:

$$\hat{\mathcal{H}} = -\tfrac{1}{2}\sum_{l,m} J_{lm}\hat{\mathbf{s}}_l\hat{\mathbf{s}}_m + |K|\,\mu^2\sum_l (s_l^z)^2 - \mu H\sum_l s_l^z. \tag{27}$$

In the ground state, when the moment is uniform, the energy is

$$E_0 = -\tfrac{1}{2}\sum_{l,m} J_{lm}s^2 + |K|\,\mu^2 s^2 N\cos^2\theta - \mu H s N\cos\theta \tag{28}$$

(θ is the angle between the z axis and the equilibrium moment).

As a result of minimization of E_0 with respect to θ, we obtain the condition that relates θ to H:

$$\cos\theta = \frac{H}{H_A} \quad \text{for } H \leqslant H_A, \tag{29}$$

$$\sin\theta = 0 \quad \text{for } H \geqslant H_A, \tag{30}$$

where the anisotropy field $H_A = 2\,|K|\,\mu s$.

On transforming to a primed system of coordinates in the same way as in the case of a positive anisotropy constant, and on repeating the same Hamiltonian-diagonalization procedure as for $K > 0$, we get

$$\mathcal{H} = \mathcal{H}_0 + \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} c_{\mathbf{k}}^{+} c_{\mathbf{k}}, \tag{31}$$

where $\epsilon_{\mathbf{k}}$ is determined, as before, by formula (15), but with the difference that $A_{\mathbf{k}}$ and B are now equal to:

$$A_{\mathbf{k}} = \begin{cases} Js(ka)^2 + \dfrac{\mu H_A}{2}\left[1 - \left(\dfrac{H}{H_A}\right)^2\right] & (H \leqslant H_A), \\ Js(ka)^2 + \mu(H - H_A) & (H \geqslant H_A), \end{cases}$$

$$B = \begin{cases} \dfrac{\mu H_A}{2}\left[\left(\dfrac{H}{H_A}\right)^2 - 1\right] & (H \leqslant H_A), \\ 0 & (H \geqslant H_A). \end{cases} \tag{32}$$

The mean moment along the z axis is determined by the equations

$$M_z = M_0\frac{H}{H_A} - \frac{H}{H_A}\frac{\mu}{4\pi^2}\int_0^\infty \frac{A_{\mathbf{k}} - \varepsilon_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}} k^2\,dk - \frac{H}{H_A}\frac{\mu}{2\pi^2}\int_0^\infty \frac{A_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}}\frac{k^2\,dk}{e^{\varepsilon_{\mathbf{k}}/T} - 1} \quad (H \leqslant H_A);$$

$$M_z = M_0 - \frac{\mu}{2\pi^2}\int_0^\infty \frac{k^2\,dk}{e^{\varepsilon_{\mathbf{k}}/T} - 1} \qquad (H \geqslant H_A). \tag{33}$$

The magnetic susceptibility at $T = 0$, in contrast to the case of a positive anisotropy constant, has at the point $H = H_A$ a finite discontinuity $\Delta\chi = M_0/H_A$.

In closing, we take this occasion to express our sincere thanks to M. I. Kaganov for helpful discussions.

[1] E. A. Turov, Fizicheskie svoĭstva magnitouporyadochennykh kristallov (Physical Properties of Magnetically Ordered Crystals), Izd. AN SSSR, 1963 [translation: Academic Press, New York, 1965].

[2] A. I. Akhiezer, V. G. Bar'yakhtar, and M. I. Kaganov, Usp. Fiz. Nauk 71, 533 (1960) and 72, 3 (1960) [Sov. Phys.-Usp. 3, 567 and 661 (1961)].

[3] T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).

[4] M. I. Kaganov and V. M. Tsukernik, Zh. Eksp. Teor. Fiz. 34, 1610 (1958) [Sov. Phys.-JETP 7, 1107 (1958)].

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