ON THE THEORY OF WEAK FERROMAGNETISM

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We have investigated a number of properties of weak ferromagnetics at low temperatures using Dzyaloshinskiĭ's theory which considers weak ferromagnetism to be the consequence of the magnetic symmetry of antiferromagnetic crystals of a well defined magnetic structure. We consider the case of "transverse" and of "longitudinal" weak ferromagnetism (in the first case the spontaneous magnetic moment is perpendicular, and in the second case parallel to the antiferromagnetic axis). We have evaluated the spin wave energy, the temperature dependence of the magnetization and its field dependence, and the spin part of the heat capacity.

1. To explain the nature of the weak ferromagnetism observed in $MnCO_3$ and $CoCO_3$, Borovik-Romanov and Orlova[1] suggested a model of ferromagnetism in which the directions of the magnetizations of the sublattices were not strictly antiparallel, but were rotated over a small angle with respect to one another. Dzyaloshinskiĭ[2] gave a group-theoretical justification of the possibility of such a magnetic state. He showed that weak ferromagnetism of compounds of the type α-Fe_2O_3 and $MnCO_3$ is a natural property of these magnetic substances in those states where the spins of the magnetic ions lie in the (111) planes and that the resulting spontaneous magnetic moment in that case is perpendicular to the antiferromagnetic axis ("transverse" weak ferromagnetism).

In the quoted paper by Borovik-Romanov and Orlova another possibility for weak ferromagnetism was also pointed out, which is produced by an incomplete compensation of the strictly antiparallel directed magnetic moments of the sublattices, due to a difference in their g-factors. This can, for instance, occur because the sublattices consist of magnetic ions (atoms) of different elements (in such a way that $N_1S_1 = N_2S_2$, but $g_1 \neq g_2$, where N_1, S_1, g_1 and N_2, S_2, g_2 are the number of ions, their spins and g-factors of the first and the second sublattice, respectively). Moreover, even for identical ions the g-factors may turn out to be different, if they occupy non-equivalent positions in the lattice so that the sites of the different sublattices have different environments. In that case, the resulting spontaneous magnetic moment of the crystal is parallel to the antiferromagnetic axis ("longitudinal" weak ferromagnetism).

Using Landau's theory of phase transitions and the most general expression for the magnetic energy which was allowed by the crystal symmetry, Dzyaloshinskiĭ investigated the behavior of weak ferromagnetics near the Curie point in its dependence on temperature and magnetic field. It is of interest to perform a similar analysis in the low temperature region where one can apply the spin wave approximation. In the present paper a first attempt in this direction is made.

2. We shall evaluate the energy of spin waves for rhombohedral crystals of the α-Fe_2O_3 and $MnCO_3$ type which are "transverse" weak ferromagnetics.

We shall describe the magnetic properties of the crystal with the aid of two magnetic moment densities $\mathbf{M}_1(\mathbf{r})$ and $\mathbf{M}_2(\mathbf{r})$ of the two magnetic sublattices respectively, and we shall assume that

$$\mathbf{M}_1^2(\mathbf{r}) = \mathbf{M}_2^2(\mathbf{r}) = \mathbf{M}_0^2 = \text{const } [3].$$

One can then write the energy density depending on the distribution of the magnetic moments in space up to terms of the second order in the direction cosines of $\mathbf{M}_1$ and $\mathbf{M}_2$ in the following form, following Dzyaloshinskiĭ[2] (see also reference 3):

$$\mathcal{H} = (A/M_0^2)\, M_{1\alpha}M_{2\alpha} + (B/2M_0^2)(M_{1z}^2 + M_{2z}^2) + (B_1/M_0^2)\, M_{1z}M_{2z} + (D/M_0^2)(M_{1x}M_{2y} - M_{2x}M_{1y}) + (C_1/M_0^2)\,\nabla M_{1\alpha}\nabla M_{2\alpha} - (\mathbf{M}_1 + \mathbf{M}_2)\,\mathbf{H} + (C/2M_0^2)(\nabla M_{1\alpha}\nabla M_{1\alpha} + \nabla M_{2\alpha}\nabla M_{2\alpha}) \quad (1)$$

($\alpha \equiv$ x, y, z; one sums over repeated indexes).

The first term here is the exchange energy between the sublattices, and the second and third terms are the usual magnetic anisotropy energy of a uniaxial crystal (trigonal axis along the z axis). The fourth term gives the magnetic energy typical for crystals of the symmetry considered here which is responsible for the weak ferromagnetism and which was taken into account for the first time by Dzyaloshinskiĭ using a correct analysis of the symmetry properties of magnetic crystals. The fifth and sixth terms take into account the change in the exchange energy produced by inhomogeneities of the magnetization where we have for the sake of simplicity only retained the isotropic part of this energy, while the last term is, finally, the energy of the magnetic substance in the external field **H**. A, B, B_1, D, C, and C_1 are quantities of the dimensions of an energy which enter as some phenomenological parameters.

The minimization of expression (1) shows that if $A > 0$, $B - B_1 > 0$, and $D > 0$ and with the field **H** in the basic XY plane the ground state of the system corresponds to those uniform distributions $\mathbf{M}_{01}$ and $\mathbf{M}_{02}$ of the magnetic moments for which they are situated in the same plane, almost antiparallel to one another, each deviating from the antiferromagnetic axis by a small angle θ such that

$$\sin\theta \approx (D + M_0 H)/2A \tag{2}$$

($\sin\theta$ is determined by the ratio of the magnetic energy to the exchange energy and θ is thus, indeed, a very small angle). In view of the fact that the initial Hamiltonian (1) does not take the magnetic anisotropy in the basic plane into account, the position of the antiferromagnetic axis is fixed by the external field **H** which is perpendicular to that axis and parallel to the resulting magnetic moment. The magnitude of the latter is, according to (2) at $T = 0°K$ equal to

$$M(0, H) = 2M_0 (H_D + H)/H_E, \tag{2'}$$

where $H_E = 2A/M_0$ is the effective field of the exchange forces (of the order of $10^6 - 10^7$ Oe), and $H_D = D/M_0$ some internal magnetic effective field (of the order of $10^3 - 10^4$ Oe) caused by the presence of the spontaneous magnetic moment at $H = 0$ which may be called the Dzyaloshinskiĭ field. The spontaneous magnetization $M_S(0) = 2M_0H_0/H_E$ is $0.01 - 1\%$ of the nominal one ($2M_0$).

Considering, further, weak oscillations $\Delta\mathbf{M}_j = \mathbf{M}_j - \mathbf{M}_{0j}$ ($j = 1, 2$) of the magnetic densities around their values corresponding to the ground state, one can by the usual means[3,4] obtain the energy of two kinds of spin waves

$$E_k^{(1)} = \{\mu^2 (H_D + H) H + I^2 k^2\}^{1/2}; \tag{3}$$

$$E_k^{(2)} = \{\mu^2 [H_0^2 + H_D (H_D + H)] + I^2 k^2\}^{1/2}, \tag{4}$$

where k is the spin wave vector, $\mu = ge\hbar/2mc$, $I^2 = 2A(C - C_1)\mu^2/M_0^2$, and $H_0 = \sqrt{2A(B - B_1)}/M_0$ is a third characteristic field the magnitude of which (of the order of $10^4 - 10^5$ Oe) is the geometric mean of the exchange forces field and the magnetic anisotropy field.

3. Using Eqs. (3) and (4) for the energy of the spin waves one can easily evaluate the temperature dependence of the magnetization. A standard calculation[3] gives

$$M(T, H) = M_s(T) + \chi(T) H, \tag{5}$$

for the temperature range $\mu H_0 \ll \kappa T \ll \kappa\Theta_C$ (Θ_C: Curie temperature, κ: Boltzmann's constant), where the spontaneous magnetization is*

$$M_s(T) = (2M_0 H_D/H_E)(1 - \alpha T^2),$$

$$\alpha = \mu^2 H_E \varkappa^2 / 24 M_0 I^3, \tag{6}$$

and the susceptibility

$$\chi(T) = (2M_0/H_E)(1 - \alpha T^2). \tag{7}$$

Borovik-Romanov and Orlova[1] obtained for $MnCO_3$ and $CoCO_3$ a two-term equation similar to (5) for the temperature and field dependence of the magnetization. As far as the specific form of the dependence of M_S and χ on the temperature is concerned one can for the moment only note the agreement of the theory with experiments as far as an increase of both quantities with a decrease of temperature is concerned and a tendency to saturation. For a more detailed verification of the theory it is necessary to extend the measurements to the region of very low temperatures (provided, of course, that there is in these substances no low-temperature transformation from the state of weak ferromagnetism to a truly antiferromagnetic state, such as, for instance, occurs in the case of hematite).

4. We shll begin the consideration of the "longitudinal" weak ferromagnetism with a discussion of the general case of a ferromagnetic with two magnetic sublattices, the magnetizations of which are antiparallel to one another in the ground state while $M_{01} \neq M_{02}$. The Hamiltonian of such a system is of the form [compare with Eq. (1)]

$$\mathcal{H} = (A_{12}/M_{01}M_{02}) M_{1\alpha} M_{2\alpha} + (C_{12}/M_{01}M_{02}) \nabla M_{1\alpha} \nabla M_{2\alpha} + (C_{11}/2M_{01}^2) \nabla M_{1\alpha} \nabla M_{1\alpha} + (C_{22}/2M_{02}^2) \nabla M_{2\alpha} \nabla M_{2\alpha} - (\mathbf{M}_1 + \mathbf{M}_2)\mathbf{H}. \tag{8}$$

*If H_0 is so large that $\mu H_0 \gg \varkappa T \gg \mu H, \mu H_D$, one must replace the coefficient α by $\alpha/2$ in Eq. (6).

if the anisotropy forces are not taken into account. The resulting magnetization in the ground state is equal to $M_{01} - M_{02}$ (if $A_{12} > 0$ and $M_{01} > M_{02}$) and directed along the field H. The spectrum of the eigenoscillations is given by the expression

$$E_k^{(1,2)} = \sqrt{(\varepsilon_1 + \varepsilon_2)^2/4 - \beta^2} \pm (\varepsilon_1 - \varepsilon_2)/2, \tag{9}$$

where

$$\varepsilon_1 = (\mu_1/M_{01})(A_{12} + C_{11}k^2) + \mu_1 H,$$

$$\varepsilon_2 = (\mu_2/M_{02})(A_{12} + C_{22}k^2) - \mu_2 H,$$

$$\beta = (\mu_1\mu_2/M_{01}M_{02})^{1/2}(A_{12} + C_{12}k^2).$$

One sees easily that the necessary and sufficient condition that the spin wave energy have the linear dispersion law ($E_k \sim k$ if $H = 0$) characteristic for antiferromagnetics is the equation

$$M_{01}/\mu_1 = M_{02}/\mu_2. \tag{10}$$

This equation allows a spontaneous magnetization to be present:

$$M_s(0) = M_{01} - M_{02} = \delta M_0,$$

where

$$\delta = \Delta\mu/\mu, \ \Delta\mu = (\mu_1 - \mu_2)/2, \ \mu = (\mu_1 + \mu_2)/2$$
$$\text{and } M_0 = (M_{01} + M_{02})/2.$$

We are here thus dealing with spin antiferromagnetism since, according to (10), $N_1S_1 = N_2S_2$, and with weak "longitudinal" ferromagnetism produced by a difference in g-factors of the magnetic ions of the different sublattices. The quantity δ which determines the possible value of the spontaneous magnetization of such hypothetical weak ferromagnetics can, generally speaking, vary within wide limits compared to the usual ferromagnetics; if we use the observed differences in g-factors for magnetic ions (atoms) of different elements or of one element, but in different compounds, we have $\delta \sim 10^{-3} - 10^{-1}$.

If for the sake of simplicity we restrict ourselves to the case $C_{11} = C_{22}$ we can write the energy of the spin waves (9) of a "longitudinal" weak ferromagnetic, taking (12) into account, in the form

$$E_k^{(1,2)} = \{\mu^2(\delta H_E H + \delta^2 H^2) + I^2k^2\}^{1/2} \pm \mu H. \tag{11}$$

We note that the states considered by us, in which the antiferromagnetic axis is parallel to **H**, are stable only in the range $H \lesssim \delta H_E$ for the field. This range of stability is changed if we take the magnetic anisotropy energy into account in (8).*

*We shall not write down here the unwieldy equations for the spin wave energy taking magnetic anisotropy into account, since the latter does not change the conclusions which interest us in the following.

The evaluation of the temperature dependence of the magnetization (for $\kappa T \gg \mu\sqrt{\delta H_E H}$) gives again an expression of the form (5) where now, however,

$$M_s(T) = \delta M_0[1 - \mu^2 H_E(\varkappa T)^2/12M_0 I^3]; \tag{12}$$

$$\chi(T) = \mu^2(\varkappa T)^2/3I^3. \tag{13}$$

It is characteristic that that part of the magnetization which depends on the temperature $\Delta M(T, H) = -\mu^2(\kappa T)^2(\delta H_E - 4H)/12I^3$ must change its sign at $H = \delta H_E/4$.

5. On the basis of the calculations reported one can reach the following conclusions:

a) Both for "transverse" and for "longitudinal" weak ferromagnetics the spontaneous magnetic moment changes with temperature not according to a $T^{3/2}$ law, characteristic for ordinary ferromagnetics* but to a T^2 law.

b) The magnetic susceptibility $\chi(T)$ in a field H directed along the spontaneous magnetic moment is for "transverse" weak ferromagnetics the same as the usual perpendicular susceptibility of an antiferromagnetic and for "longitudinal" weak ferromagnetics as the usual parallel susceptibility of an antiferromagnetic [compare Eqs. (7) and (13) for $\chi(T)$ with the corresponding formulae for $\chi_\perp$ and $\chi_\parallel$ in the author's reference 6]. This fact can be one of the indications by which one can easily distinguish "transverse" weak ferromagnetism from "longitudinal."

c) Since for all weak ferromagnetics the spin wave energy obeys a linear dispersion law, namely, $E_k = Ik$ (for $\kappa T \gg \Delta E_0$ where ΔE_0 is the energy gap for the excitation of spin waves), their spin heat capacity C_S is proportional to T^3, as in antiferromagnetics. The corresponding expressions for C_S were, for instance, given in the papers by Kaganov and Tsukernik[4] and by the present author.[6]

To check the results of the present paper, it is necessary to have additional experimental data on the magnetization, heat capacity and magnetic resonance in known weak ferromagnetics, preferably on single crystal samples and at temperatures, low compared to the Curie temperature.

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*Vonsovskii and Seidov[5] obtained a T^2 law for ferromagnetic ferrites, thenks to the fact that the problem of two magnetic sublattices with $N_1S_1 \neq N_2S_2$ and $g_1 = g_2$ was in fact replaced by them by the problem with $N_1S_1 = N_2S_2$ and $g_1 \neq g_2$, which is not equivalent to it.

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235