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Magnetization processes of free- and fixed-single-crystal antiferromagnets

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A simple two-sublattice model is used to analyze the magnetization process of anisotropic antiferromagnets at $T=0$. The magnetization is calculated for samples free to be oriented by the applied field (free single crystals or powders consisting of single-grain particles). We find that the anisotropy does not necessarily lead to a nonlinear $M(B)$ relation, but that the magnetic isotherms are broken into two linear sections separated by a discontinuity. The difference between the slopes in the two regions enables a determination of the anisotropy constant K_1 . The results will be discussed in relation to our findings for the ferrimagnetic case and to the behavior of oriented antiferromagnetic crystals. © 1996 American Institute of Physics. [S0021-8979(96)00108-0]

The study of magnetization processes in two-sublattice systems is mostly conducted with reference to rare-earth—transition-metal compounds. Such compounds are composed of two very dissimilar sublattices. The ferrimagnetic case, with antiferromagnetic intersublattice coupling, is relevant to the compounds formed with heavy-rare-earth elements. These compounds have been studied extensively in recent years because of their relationship to materials used for several technological applications, including permanent magnets, magneto-optical recording, and magnetoelastic transducer devices.

Model calculations of the magnetization processes in two-sublattice ferrimagnets have been based on a phenomenological free-energy expression consisting of the anisotropy energies of both sublattices, the energy of the interaction between the sublattices and the Zeeman energy.¹ Such analyses implicitly include the case of antiferromagnetism, where the anisotropy parameters and the magnetization of the two sublattices are identical. Such parameter values being far removed from those occurring in ferrimagnets of practical interest, this case has received little attention so far. The purpose of the present paper is to reconsider the results on ferrimagnets with this special case in mind and to point out that the magnetization of field-oriented (or free-powder) samples of antiferromagnets with uniaxial sublattice anisotropy show some general features not commonly observed in ferrimagnets. Many of such antiferromagnetic rare-earth compounds have been reported in Ref. 2.

The interest in magnetization curves of field-oriented single-crystalline samples or powders consisting of single-crystal particles arose from the realization that such curves may turn out to be very simple, consisting of three straight sections. The first one of these, stretching to a first critical field B_{cr1} , is horizontal at the value of the spontaneous magnetization $|M_1 - M_2|$. This is followed by a section where the magnetization is proportional to the field, $M = n_{12}^{-1}B$, the slope being equal to the inverse of the intersublattice coupling constant. When the magnetization reaches its saturation value $M_1 + M_2$ at a second critical field B_{cr2} , it becomes again independent of the field. This simple behavior has been shown to prevail in two cases: (1) if at least one of the

sublattices is isotropic and (2) if both sublattices have planar (easy-plane) anisotropies, with no anisotropy in the plane. Clearly, case (1) is irrelevant to anisotropic antiferromagnets and case (2) implies that easy-plane antiferromagnets show the simple magnetization curves known from the ferrimagnet case. Since $M_1 - M_2 = 0$ for antiferromagnets, the first one of the three straight sections described above is absent; the bending of the two sublattice magnetizations towards each other begins at $B_{cr1} = 0$, the linear increase of the magnetization with a slope n_{12}^{-1} stretching to $M = M_1 + M_2$, which is reached at $B_{cr2} = n_{12}(M_1 + M_2)$. This being a very straightforward application of the results known for ferrimagnets, we shall concentrate on antiferromagnets with easy-axis anisotropy in what follows, which will be shown to have some interesting features.

The simplest anisotropic two-sublattice magnets, when they are free to be oriented by the field, can be described by the free energy

$$E = K_1^{(1)} \sin^2 \vartheta_1 + K_1^{(2)} \sin^2 \vartheta_2 + n_{12} M_1 M_2 \cos \alpha - BM, \quad (1)$$

where the first two terms stand for the anisotropy energy, with ϑ_i the angle between the magnetization vector $\mathbf{M}_i$ and the c axis, α is the angle between the magnetization vectors, and M is the magnitude of the total magnetization vector $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$, which is supposed to point into the direction of the applied field $\mathbf{B}$ throughout the magnetization process. It has been shown¹ that the bending of the two magnetization vectors towards each other always takes place within a plane containing the c axis and therefore two angular variables suffice to characterize any state occurring in the process. It turns out that the symmetry properties of an antiferromagnet, $M_1 = M_2 = M_0$ and $K_1^{(1)} = K_1^{(2)} = K_1$, can be best exploited if the free energy is written in terms of $\vartheta = (\vartheta_1 + \vartheta_2)/2$, the angle between the total magnetization vector and the c axis, and $\alpha = \vartheta_2 - \vartheta_1$, the angle between the sublattice magnetizations:

$$E(\vartheta, \alpha) = 2K_1 \left(\sin^2 \vartheta \cos^2 \frac{\alpha}{2} + \cos^2 \vartheta \sin^2 \frac{\alpha}{2} \right) + n_{12} M_0^2 \cos \alpha - 2BM_0 \cos \frac{\alpha}{2}. \quad (2)$$

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FIG. 1. (a) Field dependence of the free energy for a free-single-crystal antiferromagnet with easy-axis anisotropy. (b) Field dependence of magnetization. $T=0$ K, $B_{\text{sat}}(0) > B_x$.

To find the minima of the function (2), we first differentiate with respect to ϑ to find

$$\frac{\partial E}{\partial \vartheta} = 2K_1 \sin 2\vartheta \cos \alpha; \quad \frac{\partial^2 E}{\partial \vartheta^2} = 4K_1 \cos 2\vartheta \cos \alpha. \quad (3)$$

The condition $\partial E / \partial \vartheta = 0$ is satisfied at $\alpha = \pi/2$, but this is not a minimum since in this case $\partial^2 E / \partial \vartheta^2$ (as well as all further derivatives) vanishes. The other condition for a vanishing first derivative is $\sin 2\vartheta = 0$. There are two cases of interest, $\vartheta = 0$ and $\vartheta = \pi/2$, where the necessary condition of a minimum, $\partial^2 E / \partial \vartheta^2 > 0$, will be satisfied for $\alpha < \pi/2$ and $\alpha > \pi/2$, respectively. These findings accord with the intuitive notion of uniaxial antiferromagnets: at low fields, where the magnetization vectors are still close to the antiparallel configuration, the $\mathbf{B} \perp c$ ($\vartheta = \pi/2$) configuration is favorable, whereas close to the forced ferromagnetic alignment $\mathbf{B} \parallel c$ ($\vartheta = 0$) gives the lowest anisotropy energy.

Though it is clear that the two conditions found above correspond to two distinct regions, it is instructive to derive the free energy as a function of the magnetic field for both. From Eq. (2) we find for $\vartheta = \pi/2$,

$$E\left(\frac{\pi}{2}, \alpha\right) = 2K_1 \cos^2 \frac{\alpha}{2} + n_{12}M_0^2 \cos \alpha - 2BM_0 \cos \frac{\alpha}{2} \quad (4)$$

and for $\vartheta = 0$,

$$E(0, \alpha) = 2K_1 \sin^2 \frac{\alpha}{2} + n_{12}M_0^2 \cos \alpha - 2BM_0 \cos \frac{\alpha}{2}. \quad (5)$$

The conditions for energy minima, $dE/d\alpha = 0$, give in both cases an explicit expression for $\cos \alpha/2$ ($\sin \alpha/2 \neq 0$) which, substituted back into Eqs. (4) and (5) give the desired functions of the applied field: for $\vartheta = \pi/2$,

$$E\left(\frac{\pi}{2}, B\right) = -n_{12}M_0^2 - \frac{B^2}{2(n_{12} + K_1/M_0^2)} \quad (6)$$

and for $\vartheta = 0$,

$$E(0, B) = 2K_1 - n_{12}M_0^2 - \frac{B^2}{2(n_{12} - K_1/M_0^2)}. \quad (7)$$

Through the relation $M = \partial E / \partial B$ both of these equations imply linearly increasing magnetizations, but this of course cannot prevail beyond saturation where, with $\alpha = 0$, Eqs. (4) and (5) give

$$E_{\text{sat}}\left(\frac{\pi}{2}, B\right) = 2K_1 + n_{12}M_0^2 - 2BM_0, \quad (8)$$

$$E_{\text{sat}}(0, B) = n_{12}M_0^2 - 2BM_0. \quad (9)$$

Equating the respective energies given by Eqs. (6), (7) and by Eqs. (8) and (9) gives the field values where saturation sets in. In both cases we find only one such value,

$$B_{\text{sat}}\left(\vartheta = \frac{\pi}{2}\right) = 2M_0\left(n_{12} + \frac{K_1}{M_0^2}\right), \quad (10)$$

$$B_{\text{sat}}(\vartheta = 0) = 2M_0\left(n_{12} - \frac{K_1}{M_0^2}\right) \quad (11)$$

indicating that the straight lines given by Eqs. (8) and (9) are tangent to the parabolae (6) and (7), as illustrated by Fig. 1(a).

Having constructed the free-energy curves, it is now easy to identify the lowest-energy track, which will be followed in the magnetization process. Clearly, up to $B = B_x$, where the two parabolae cross, $\vartheta = \pi/2$ and beyond this field $\vartheta = 0$. Setting the energies given by Eqs. (6) and (7) equal, we find

$$B_x^2 = 2M_0^2\left[n_{12}^2 - \left(\frac{K_1}{M_0^2}\right)^2\right]; \quad (3K_1 \leq M_0^2 n_{12}). \quad (12)$$

The condition satisfied by this equation will be discussed below. The slopes of the two curves being different at B_x , there must be a discontinuity in the magnetization,

FIG. 2. (a) Field dependence of the free energy for a free-single-crystal antiferromagnet with easy-axis anisotropy. (b) Field dependence of magnetization. $T=0$ K, $B_{\text{sat}}(0) < B_x$.

FIG. 3. Field dependence of the free energy and magnetization for a fixed-single-crystal antiferromagnet with easy-axis anisotropy. (a) The applied field is along the hard-magnetization direction. (b) The applied field is along the easy-magnetization direction.

$$\Delta M = 2\sqrt{2}M_0 \sqrt{(K_1/M_0^2)^2/[n_{12}^2 - (K_1/M_0^2)^2]}. \quad (13)$$

Figure 1(b) shows the magnetization curve derived from the solid free-energy curve in Fig. 1(a). The magnetization will jump directly to its saturation value whenever $B_{\text{sat}}(\vartheta=0) < B_x$. Using Eqs. (11) and (12), we find that this will be the case when $3K_1 > n_{12}M_0^2$. Under this condition, Eq. (12) has to be replaced by the expression found after equating the energies given by Eqs. (6) and (9)

$$B_x = 2M_0(n_{12} + K_1/M_0^2)(1 - \sqrt{K_1/[n_{12}M_0^2 + K_1]}), \quad (14)$$

$$3K_1 \geq n_{12}M_0^2.$$

Figure 2 shows the free-energy and magnetization curves for such a case.

The linear behavior of the magnetization seen in Figs. 1 and 2 is a consequence of the simple form of the anisotropy energy assumed in Eq. (1). However, the occurrence of a jump in the magnetization is a general feature of easy-axis antiferromagnets, as can be seen from the following argument.

Replacing the lowest-order expression $K_1 \sin^2 \vartheta$ by a more general form $E_{\text{an}}(\vartheta)$ of the sublattice anisotropy energy will not prevent the crossing of the $E(\pi/2, B)$ and $E(0, B)$ curves. Since $E_{\text{an}}(0) < E_{\text{an}}(\pi/2)$ must hold to ensure easy-axis anisotropy, it follows that $E(\pi/2, 0) - E(0, 0) = 2[E_{\text{an}}(0) - E_{\text{an}}(\pi/2)] < 0$, whereas at saturation (in the

forced ferromagnetic state) $E_{\text{sat}}(\pi/2, B) - E_{\text{sat}}(0, B) = 2[E_{\text{an}}(\pi/2) - E_{\text{an}}(0)] > 0$. The form of the function $E_{\text{an}}(\vartheta)$ is thus seen to influence the shape of the curved parts of the free-energy functions, but the order of the energy values at zero field and high fields ($B > B_{\text{sat}}$) will remain inverted, making an intercept with different slopes and hence a jump in the magnetization inevitable.

The discontinuity in the magnetization is reminiscent of first-order phase transitions, but it should be born in mind that this transition is related to the additional degrees of freedom introduced by the free rotation of the crystals. Indeed, switching from the $\vartheta = \pi/2$ curve to the $\vartheta = 0$ one in Figs. 1 and 2 signifies a rotation of the sample by 90° . The discontinuous changes (spin-flop transitions) known to occur in oriented antiferromagnets are of a different nature. Preliminary experimental results have been reported in Ref. 3.

The free-energy functions (6)–(9) are relevant to the magnetization process in samples fixed at $\mathbf{B} \perp c$ and $\mathbf{B} \parallel c$ orientations. If $\mathbf{B} \perp c$, the bending process traces the $E(\pi/2, B)$ curve to $B_{\text{sat}}(\vartheta = \pi/2)$ and the $E_{\text{sat}}(\pi/2, B)$ line thereafter. The magnetization increases continuously up to saturation [Fig. 3(a)]. If $\mathbf{B} \parallel c$, bending does not begin at $B = 0$, because in weak fields the Zeeman energy cannot overcome the anisotropy. The sublattice magnetizations remain in the c direction, fully compensated ($M = 0$), until the point is reached (at B_f), where the $E(0, B)$ curve falls below $E(\pi/2, 0)$ [Fig. 3(b)]. The transition taking place at this point is not necessarily to a state with $\alpha < \pi/2$, which is one of the features distinguishing it from what takes place in freely rotating samples. Another related feature is the distinct value of B_f , which follows from the condition $E(\pi/2, 0) = E(0, B_f)$: $B_f^2 = 4K_1(n_{12} - K_1/M_0^2)$, provided $2K_1 \leq n_{12}M_0^2$. The limitation of K_1 to low values comes from the requirement that B_f be less than $B_{\text{sat}}(\vartheta = 0)$. If that is not the case, the spin-flop will be total, leading to saturation, and B_f is to be determined from the condition $E(\pi/2, 0) = E_{\text{sat}}(0, B_f)$, which gives $B_f = n_{12}M_0$, provided $2K_1 \geq n_{12}M_0^2$. Comparing these results with Eqs. (12) and (14) it clear that B_x and B_f mark the occurrence of transitions of different nature.

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