Law of approach to saturation for polycrystalline ferromagnets: Remanent initial state

J. F. Herbst and F. E. Pinkerton
Physics and Physical Chemistry Department, General Motors Research and Development Center, 30500 Mound Road, Warren, Michigan 48090-9055
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We consider the approach to saturation for cubic and uniaxial polycrystalline, single-domain ferromagnets from the remanent state. For both cubic {111}- and {100}-easy systems we find that the coefficient β in the law $M(H) \underset{H\to\infty}{\approx} M(1-\beta K^2/M^2H^2)$, where M is the saturation magnetization and K is the principal anisotropy constant, departs from the well-known result $\beta=8/105$ obtained for an initially isotropic moment distribution. The coefficients for the two easy-axis systems become distinct, and we calculate the dependence of β on the angle between the primary magnetic field which established the remanent state and the second saturating field H. For uniaxial systems we show that $\beta=4/15$ for the remanent state, identical to its standard value for the isotropic initial state. [S0163-1829(98)08217-4]

I. INTRODUCTION

For an initially isotropic distribution of magnetic moments which can change direction only by rotating against the magnetic anisotropy, it is well known (see, for example, Refs. 1 and 2) that the magnetization $M(H)$ approaches its saturation value M in large magnetic field H according to the law

$$M(H) \underset{H\to\infty}{\approx} M(1-\beta K^2/M^2H^2), \tag{1}$$

where K is the principal anisotropy constant. If such a polycrystalline ferromagnet consists of randomly oriented, single-domain crystallites having cubic anisotropy, $\beta\equiv\beta_{\rm iso}=8/105$, while for uniaxial systems $\beta_{\rm iso}=4/15$, the subscript denoting the isotropic initial state (characteristic, for example, of a thermally demagnetized specimen). Here we point out that β departs from $\beta_{\rm iso}$ for a cubic system if the initial moment configuration is the remanent state, that is, if the magnet has already been saturated and then allowed to relax to the remanent state in which the moment of each crystallite resides along the easy axis closest to the primary saturating field direction with a positive component in that direction. We find that the coefficient $\beta_{\rm rem}$ appearing in Eq. (1) for the remanent initial state depends not only on the easy-axis preference ({111} or {100}) for cubic systems, but also on the angle ρ between the directions of the first saturating field (used to prepare the remanent state) and the second saturating field.

In the following we assume that the primary saturating field has been applied along the z direction of a standard Cartesian coordinate system and then removed, forming the remanent state. We specify the magnetization $\mathbf{M}$ and easy direction $\hat{n}$ of each crystallite as well as the (second) applied field $\mathbf{H}$ in terms of polar and azimuthal angle sets referred to the same global coordinate system:

$$\mathbf{M}=M(\sin\theta\cos\phi\hat{x}+\sin\theta\sin\phi\hat{y}+\cos\theta\hat{z}), \tag{2}$$

$$\hat{n}=\sin\Theta\cos\Phi\,\hat{x}+\sin\Theta\sin\Phi\hat{y}+\cos\Theta\hat{z}, \tag{3}$$

$$\mathbf{H}=H(\sin\rho\cos\sigma\hat{x}+\sin\rho\sin\sigma\hat{y}+\cos\rho\hat{z}). \tag{4}$$

The coefficient β is obtained by minimizing (with respect to θ and ϕ) the total energy of each crystallite

$$E=E_{\rm mca}-\mathbf{M}\cdot\mathbf{H}=E_{\rm mca}-MH\cos\Delta, \tag{5}$$

where $E_{\rm mca}=E_{\rm mca}(\theta,\phi)$ is the magnetocrystalline anisotropy energy of a single crystallite and Δ is the angle between $\mathbf{M}$ and $\mathbf{H}$, for small Δ (i.e., large H). This yields $M(H)=M\cos\Delta$ in the form of Eq. (1) with

$$\beta=\frac{1}{2K^2}|\nabla E_{\rm mca}(\rho,\sigma)|^2=\frac{1}{2K^2}\left\{\left[\frac{\partial E_{\rm mca}(\rho,\sigma)}{\partial\theta}\right]^2+\frac{1}{\sin^2\rho}\left[\frac{\partial E_{\rm mca}(\rho,\sigma)}{\partial\phi}\right]^2\right\}, \tag{6}$$

in which the gradient of $E_{\rm mca}$ is evaluated along the field direction ($\theta=\rho$, $\phi=\sigma$). For an assembly of moments β must be obtained from an appropriate average over the moment distribution, a procedure we develop on a case-by-case basis below. Sections II and III describe the calculation of $\beta_{\rm rem}(\rho)$ for cubic {111}- and {100}-easy systems, respectively, and in Sec. IV we demonstrate that $\beta_{\rm rem}=\beta_{\rm iso}$ for uniaxial anisotropy. Concluding remarks are made in Sec. V.

II. CUBIC ANISOTROPY: {111} EASY

We identify the locus of moments (i.e., occupied {111} directions) for a random polycrystal in the remanent state by means of the following considerations. For a given crystallite with a set of Cartesian axes fixed in it, the locus of directions nearest a particular {111} direction $\mathbf{P}$ is the octant of the unit sphere containing $\mathbf{P}$ in the crystallite coordinate frame; an octant is the appropriate region since there are eight symmetrically positioned {111} directions. Conversely, having chosen $\hat{z}$ in our global Cartesian system as the primary saturating field direction, a possible locus for the populated easy {111} directions of the entire assembly of crystallites in the remanent state is an octant centered about $\hat{z}$. Figure 1 shows the projection of one such octant in the (x,y) plane; it is-

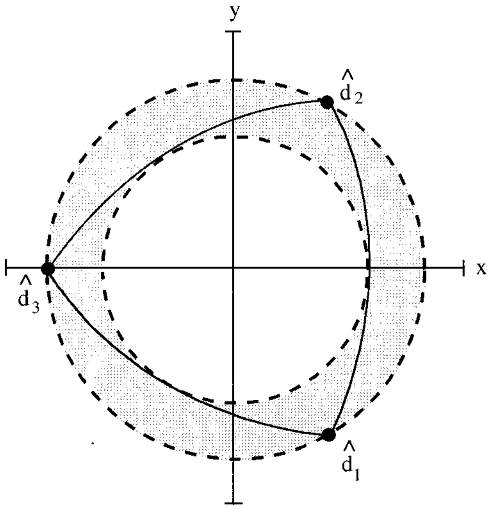

FIG. 1. Projection into the (x,y) plane of an octant of the unit sphere relevant to the {111}-easy remanent state. $\hat{z}$ (out of the paper) is the direction of the primary saturating field which generates the remanent state. Dots represent the tips of the $\hat{d}_i$ vectors specified by Eqs. (7). The dotted lines are the circles inscribing and circumscribing the octant. In the remanent state the distribution of easy directions [$g_{111}(\Theta)$ of Eq. (10c)] is constant within the inscribed circle and decreases to zero in the shaded region (swept out by rotation of the octant) between the inscribed and circumscribed circles.

defined[3] by great circle segments passing through the vertices

$$\hat{d}_1=\frac{1}{\sqrt{6}}(\hat{x}-\sqrt{3}\hat{y}+\sqrt{2}\hat{z}), \tag{7a}$$

$$\hat{d}_2=\frac{1}{\sqrt{6}}(\hat{x}+\sqrt{3}\hat{y}+\sqrt{2}\hat{z}), \tag{7b}$$

and

$$\hat{d}_3=\frac{1}{\sqrt{3}}(-\sqrt{2}\hat{x}+\hat{z}). \tag{7c}$$

The average of a function $f(\Theta,\Phi)$ over this octant can be written as

$$\langle f(\Theta,\Phi)\rangle=\frac{2}{\pi}\int_{-\pi/3}^{\pi/3}d\Phi\int_0^{\cot^{-1}(\sqrt{2}\cos\Phi)}d\Theta\,\sin\Theta\left[f(\Theta,\Phi)+f\left(\Theta,\Phi+\frac{2\pi}{3}\right)+f\left(\Theta,\Phi+\frac{4\pi}{3}\right)\right]. \tag{8}$$

(Averages, that is, integrals normalized to the area of integration over the unit sphere, are appropriate since our interest in this paper is the behavior of the magnetization rather than the total magnetic moment.) The orientation of the (x,y) plane about $\hat{z}$ is arbitrary, however; rotation of the octant in Fig. 1 by an arbitrary angle ζ about $\hat{z}$ generates an equivalent octant, so that an average over ζ must be performed in general to calculate properties of the remanent state. Manipulating the order of integration and using the fact that

$$\int_0^{2\pi}d\zeta\int_{a+\zeta}^{b+\zeta}d\Phi f(\Theta,\Phi)=\int_0^{2\pi}d\Phi(b-a)f(\Theta,\Phi) \tag{9}$$

[for $0\leqslant b-a\leqslant 2\pi$ and $f(\Theta,\Phi)$ periodic in Φ], we average $\langle f(\Theta,\Phi);\zeta\rangle$ for a given ζ over $0\leqslant\zeta\leqslant 2\pi$:[4]

$$\begin{aligned}\langle f(\Theta,\Phi)\rangle_{111}&\equiv\frac{1}{2\pi}\int_0^{2\pi}d\zeta\langle f(\Theta,\Phi);\zeta\rangle\\&=\frac{1}{2\pi}\frac{2}{\pi}\int_0^{2\pi}d\zeta\left\{\int_0^{\Theta_{\min}}d\Theta\,\sin\Theta\int_{-\pi/3+\zeta}^{\pi/3+\zeta}d\Phi+\int_{\Theta_{\min}}^{\Theta_{\max}}d\Theta\,\sin\Theta\int_{\cos^{-1}[(1/\sqrt{2})\cot\Theta]+\zeta}^{\pi/3+\zeta}d\Phi\right.\\&\quad\left.+\int_{\Theta_{\min}}^{\Theta_{\max}}d\Theta\,\sin\Theta\int_{-\pi/3+\zeta}^{-\cos^{-1}[(1/\sqrt{2})\cot\Theta]+\zeta}\right\}\left[f(\Theta,\Phi)+f\left(\Theta,\Phi+\frac{2\pi}{3}\right)+f\left(\Theta,\Phi+\frac{4\pi}{3}\right)\right]\\&\equiv\int d\Omega\,g_{111}(\Theta)f(\Theta,\Phi),\end{aligned} \tag{10a}$$

where

$$\int d\Omega\cdots\equiv\int_0^{\pi}d\Theta\,\sin\Theta\int_0^{2\pi}d\Phi\cdots \tag{10b}$$

indicates integration over the unit sphere, $\Theta_{\min}=\sin^{-1}1/\sqrt{3}$ and $\Theta_{\max}=\sin^{-1}\sqrt{2/3}$ are the polar angles, respectively, corresponding to the circles inscribing and circumscribing the octant (dotted lines in Fig. 1), and $g_{111}(\Theta)$ is defined by

$$g_{111}(\Theta)=\frac{2}{\pi}\times\begin{cases}1, & 0\leqslant\Theta\leqslant\Theta_{\min}\\ 1-\frac{3}{\pi}\cos^{-1}[(1/\sqrt{2})\cot\Theta], & \Theta_{\min}\leqslant\Theta\leqslant\Theta_{\max}\\ 0, & \Theta_{\max}\leqslant\Theta\leqslant\pi.\end{cases} \tag{10c}$$

With $f(\Theta,\Phi)=1$ it is clear that

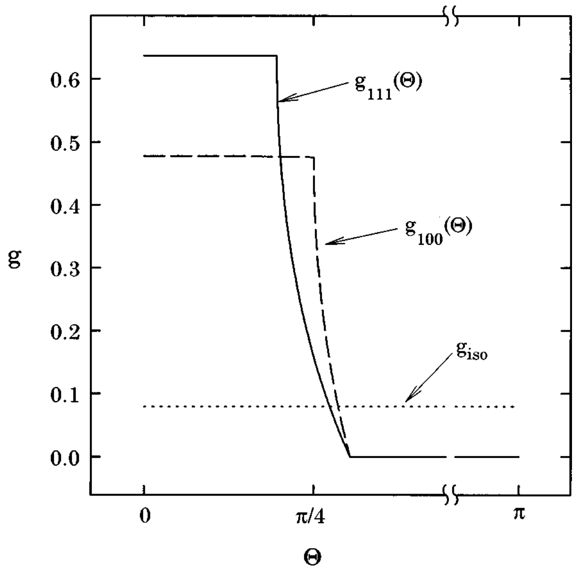

FIG. 2. Distributions $g_{111}(\Theta)$ [Eq. (10c)], $g_{100}(\Theta)$ [Eq. (27b)], and $g_{\rm iso}=1/4\pi$ of occupied easy directions in the remanent {111}, remanent {100}, and isotropic states, respectively, as functions of the polar angle Θ about the primary saturating field direction $\hat{z}$.

$$1=\int d\Omega\, g_{111}(\Theta), \tag{11}$$

so that $g_{111}(\Theta)$ can be identified as the normalized angular distribution of {111} moments about the primary saturating field direction $\hat{z}$. We see, therefore, that the locus of moments in the remanent state is not a single octant but a spherical cap centered around $\hat{z}$. As Eq. (10c) specifies, the distribution $g_{111}(\Theta)$ is azimuthally symmetric (i.e., Φ independent), constant for $\Theta\leqslant\Theta_{\rm min}$, and decreases to zero in the shaded region between the inscribed and circumscribed circles of Fig. 1; it is plotted in Fig. 2. In contrast, the distribution in the isotropic state is a constant, $g_{\rm iso}=1/4\pi$, over the entire unit sphere.

From either Eq. (8) or Eqs. (10) the component of the total magnetization along $\hat{z}$ in the remanent state ($\theta=\Theta$, $\phi=\Phi$ for each $\mathbf{M}$) is

$$\frac{M_z^{\rm tot}}{M}=\langle\cos\Theta\rangle_{111}=\langle\cos\Theta\rangle=\frac{\sqrt{3}}{2}\cong 0.8660, \tag{12a}$$

as is to be expected;[5] also,

$$\frac{M_x^{\rm tot}}{M}=\langle\sin\Theta\,\cos\Phi\rangle_{111}=\langle\sin\Theta\,\cos\Phi\rangle=0 \tag{12b}$$

and

$$\frac{M_y^{\rm tot}}{M}=\langle\sin\Theta\,\sin\Phi\rangle_{111}=\langle\sin\Theta\,\sin\Phi\rangle=0. \tag{12c}$$

The magnetocrystalline anisotropy energy of each crystallite can be expressed[6] as

$$E_{\rm mca}=-\frac{1}{2}K(\alpha_1^4+\alpha_2^4+\alpha_3^4) \tag{13}$$

with $K<0$ for {111} easy; we determine the direction cosines α_i of each $\mathbf{M}$ with respect to a set of orthogonal axes corresponding to $\hat{n}$ for that crystallite in the following way. Introducing the rotation matrix

$$R(\theta,\phi)\equiv\begin{pmatrix}\cos\theta\cos\phi & -\sin\phi & \sin\theta\cos\phi\\ \cos\theta\sin\phi & \cos\phi & \sin\theta\sin\phi\\ -\sin\theta & 0 & \cos\theta\end{pmatrix}, \tag{14}$$

we observe that $\hat{M}=R(\theta,\phi)\hat{z}$ and $\hat{n}=R(\Theta,\Phi)\hat{z}$. Now the $\hat{d}_i$ vectors of Eqs. (7) form an orthonormal set centered about $\hat{z}$, but so do the vectors $R(\xi)\hat{d}_i$, where

$$R(\xi)\equiv R(\theta=0,\phi=\xi)=\begin{pmatrix}\cos\xi & -\sin\xi & 0\\ \sin\xi & \cos\xi & 0\\ 0 & 0 & 1\end{pmatrix} \tag{15}$$

rotates a vector by an angle ξ about $\hat{z}$. For a given ξ the vectors $R(\Theta,\Phi)R(\xi)\hat{d}_i$ constitute an orthonormal set centered about $R(\Theta,\Phi)\hat{z}=\hat{n}$; orthonormality is preserved since $R(\Theta,\Phi)$ and $R(\xi)$ are proper rotation matrices. Hence, we can write

$$\begin{aligned}\alpha_i&\equiv\hat{M}\cdot R(\Theta,\Phi)R(\xi)\hat{d}_i=R(\theta,\phi)\hat{z}\cdot R(\Theta,\Phi)R(\xi)\hat{d}_i\\ &=\hat{z}\cdot R^{-1}(\theta,\phi)R(\Theta,\Phi)R(\xi)\hat{d}_i,\end{aligned} \tag{16}$$

with the understanding that an average over the arbitrary angle ξ must also be performed to provide a full polycrystalline average in determining β. Equation (16) considerably simplifies determination of the α_i; it leads to

$$\begin{aligned}E_{\rm mca}=&-\frac{1}{72}K[(a-\sqrt{3}b+\sqrt{2}c)^4\\ &+(a+\sqrt{3}b+\sqrt{2}c)^4+4(\sqrt{2}a-c)^4]\\ =&-\frac{1}{12}K[3+6c^2-7c^4+4\sqrt{2}ac(3-3c^2-4a^2)]\end{aligned} \tag{17a}$$

with

$$\begin{aligned}a\equiv a(\theta,\phi)\equiv&\sin\theta[\cos\Theta\,\cos(\phi-\Phi)\cos\xi\\ &+\sin(\phi-\Phi)\sin\xi]-\cos\theta\sin\Theta\,\cos\xi,\end{aligned} \tag{17b}$$

$$\begin{aligned}b\equiv b(\theta,\phi)\equiv&\sin\theta[-\cos\Theta\,\cos(\phi-\Phi)\sin\xi\\ &+\sin(\phi-\Phi)\cos\xi]+\cos\theta\sin\Theta\,\sin\xi,\end{aligned} \tag{17c}$$

and

$$c\equiv c(\theta,\phi)=\sin\theta\sin\Theta\,\cos(\phi-\Phi)+\cos\theta\cos\Theta, \tag{17d}$$

$a^2+b^2+c^2=1$. Using Eqs. (17) in Eq. (6), followed by averaging over $0\leqslant\xi\leqslant 2\pi$,[4] we find (after lengthy but straightforward calculation)

$$\beta_{\rm rem}^{111}=\frac{1}{18}\langle -33c^8+60c^6-36c^4+8c^2+1\rangle_{111}, \tag{18}$$

with

$$c\equiv c(\rho,\sigma)=\sin\rho\sin\Theta\,\cos(\sigma-\Phi)+\cos\rho\cos\Theta. \tag{19}$$

Performing the Φ integration of Eq. (10) removes all dependence on σ, the azimuthal angle of $\mathbf{H}$ [cf. Eq. (4)], and we obtain

$$\beta_{\mathrm{rem}}^{111}(\rho)=\frac{1}{18}\left\{\sin^8\rho\left[-\frac{212355}{128}\langle\cos^8\Theta\rangle+\frac{99099}{32}\langle\cos^6\Theta\rangle-\frac{114345}{64}\langle\cos^4\Theta\rangle+\frac{10395}{32}\langle\cos^2\Theta\rangle-\frac{1155}{128}\right]\right.$$
$$+\sin^6\rho\left[\frac{14157}{4}\langle\cos^8\Theta\rangle-\frac{13167}{2}\langle\cos^6\Theta\rangle+3780\langle\cos^4\Theta\rangle-\frac{1365}{2}\langle\cos^2\Theta\rangle+\frac{75}{4}\right]+\sin^4\rho\left[-\frac{9801}{4}\langle\cos^8\Theta\rangle\right.$$
$$\left.+4536\langle\cos^6\Theta\rangle-\frac{10395}{4}\langle\cos^4\Theta\rangle+\frac{945}{2}\langle\cos^2\Theta\rangle-\frac{27}{2}\right]+\sin^2\rho[594\langle\cos^8\Theta\rangle-1092\langle\cos^6\Theta\rangle+630\langle\cos^4\Theta\rangle$$
$$\left.-120\langle\cos^2\Theta\rangle+4]-33\langle\cos^8\Theta\rangle+60\langle\cos^6\Theta\rangle-36\langle\cos^4\Theta\rangle+8\langle\cos^2\Theta\rangle+1\right\}, \tag{20}$$

where we have omitted the 111 subscripts since the remaining averages over $\cos^n\Theta$ are independent of octant orientation. From either Eq. (8) or Eq. (10) we have

$$\langle\cos^n\Theta\rangle_{111}=\langle\cos^n\Theta\rangle=\frac{4}{n+1}\left[1-\frac{3}{\pi}\int_0^{\pi/3}d\Phi\left(\frac{\sqrt{2}\cos\Phi}{\sqrt{1+2\cos^2\Phi}}\right)^{n+1}\right], \tag{21}$$

and the required integrals can be evaluated with the help of Ref. 7:

$$\langle\cos^2\Theta\rangle_{111}=\frac{1}{3}+\frac{4}{3\pi}, \tag{22a}$$

$$\langle\cos^4\Theta\rangle_{111}=\frac{1}{5}+\frac{56}{45\pi}, \tag{22b}$$

$$\langle\cos^6\Theta\rangle_{111}=\frac{1}{7}+\frac{1012}{945\pi}, \tag{22c}$$

$$\langle\cos^8\Theta\rangle_{111}=\frac{1}{9}+\frac{7792}{8505\pi}. \tag{22d}$$

Hence, we find that the coefficient β in the law of approach to saturation from the remanent state of a {111}-easy polycrystal is

$$\beta_{\mathrm{rem}}^{111}(\rho)=\frac{8}{105}-\frac{1}{\pi}\left(\frac{32}{5103}+\frac{64}{945}\sin^2\rho-\frac{92}{189}\sin^4\rho+\frac{6484}{8505}\sin^6\rho-\frac{979}{2835}\sin^8\rho\right), \tag{23}$$

where ρ is the angle between the field which established the remanent state and the second saturating field.[8]

A consistency check on our work can be made in the following way. Averaging the right side of Eq. (18) over the entire unit sphere instead of the indicated average leads to Eq. (20) but with the $\langle\cos^n\Theta\rangle_{111}$ terms replaced by

$$\langle\cos^n\Theta\rangle_{\text{unit sphere}}=\frac{1}{4\pi}\int d\Omega\cos^n\Theta=\frac{1}{n+1}; \tag{24}$$

substitution of these into Eq. (20) yields $8/105=\beta_{\mathrm{iso}}$ independent of the direction of **H**. We thus properly recover the known result for an initially isotropic distribution of moments.

III. CUBIC ANISOTROPY: {100} EASY

Our analysis of this case proceeds in a way similar to that for the {111}-easy system in Sec. II. A specific locus for the easy directions occupied in the remanent state is a sextant of the unit sphere since there are six {100} directions. The (x,y) projection of the sextant defined by great circle segments intersecting the vertices

$$\hat{d}_1=\frac{1}{\sqrt{3}}(\hat{x}-\hat{y}+\hat{z}), \tag{25a}$$

$$\hat{d}_2=\frac{1}{\sqrt{3}}(\hat{x}+\hat{y}+\hat{z}), \tag{25b}$$

$$\hat{d}_3=\frac{1}{\sqrt{3}}(-\hat{x}+\hat{y}+\hat{z}), \tag{25c}$$

and

$$\hat{d}_4=\frac{1}{\sqrt{3}}(-\hat{x}-\hat{y}+\hat{z}) \tag{25d}$$

is shown in Fig. 3, and

$$\langle f(\Theta,\Phi)\rangle=\frac{3}{2\pi}\int_{-\pi/4}^{\pi/4}d\Phi\int_0^{\cot^{-1}(\cos\Phi)}d\Theta\sin\Theta\left[f(\Theta,\Phi)+f\left(\Theta,\Phi+\frac{\pi}{2}\right)+f(\Theta,\Phi+\pi)+f\left(\Theta,\Phi+\frac{3\pi}{2}\right)\right] \tag{26}$$

defines the average of $f(\Theta,\Phi)$ over it. In analogy to our work in Sec. II, an average over the arbitrary orientation angle ζ of the sextant about $\hat{z}$ must also be performed.[9] This leads to

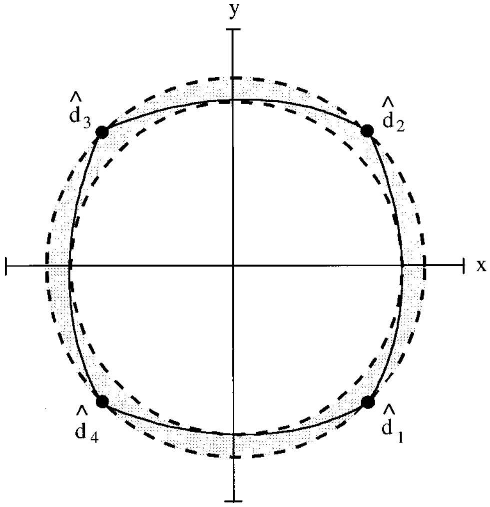

FIG. 3. Projection into the (x,y) plane of a sextant of the unit sphere relevant to the {100}-easy remanent state. $\hat{z}$ (out of the paper) is the direction of the primary saturating field. Dots represent the tips of the $\hat{d}_i$ vectors specified by Eqs. (25). The dotted lines are the circles inscribing and circumscribing the sextant. In the remanent state the distribution of easy directions [$g_{100}(\Theta)$ of Eq. (27b)] is constant within the inscribed circle and decreases to zero in the shaded region (swept out by rotation of the sextant) between the inscribed and circumscribed circles.

$$\langle f(\Theta,\Phi)\rangle_{100} \equiv \frac{1}{2\pi}\int_0^{2\pi} d\zeta \langle f(\Theta,\Phi);\zeta\rangle$$
$$\equiv \int d\Omega g_{100}(\Theta) f(\Theta,\Phi), \quad (27a)$$

where $g_{100}(\Theta)$ is the normalized angular distribution of {100} moments about the primary saturating field direction $\hat{z}$ in the remanent state:

$$g_{100}(\Theta) = \frac{3}{2\pi}\times\begin{cases} 1, & 0\leq\Theta\leq\Theta_{\min} \\ 1-\frac{4}{\pi}\cos^{-1}(\cot\Theta), & \Theta_{\min}\leq\Theta\leq\Theta_{\max} \\ 0, & \Theta_{\max}\leq\Theta\leq\pi. \end{cases} \quad (27b)$$

$\Theta_{\min}=\pi/4$ and $\Theta_{\max}=\sin^{-1}\sqrt{2/3}$ are the polar angles, respectively, corresponding to the circles inscribing and circumscribing the sextant indicated by the dotted lines in Fig. 3. The moments in the remanent state reside once more in a spherical cap centered around $\hat{z}$. The azimuthally symmetric distribution $g_{100}(\Theta)$ decreases from a constant to zero in the shaded region of Fig. 3; it is shown in Fig. 2. Using either Eq. (26) or Eq. (27) we find that the only nonzero component of the total magnetization in the remanent state is

$$\frac{M_z^{\text{tot}}}{M} = \langle\cos\Theta\rangle_{100} = \langle\cos\Theta\rangle = \frac{3}{\sqrt{2}}\left[1-\frac{2}{\pi}\tan^{-1}(\sqrt{2})\right]$$
$$\cong 0.8312, \quad (28)$$

the standard result.[10]

With appropriate direction cosines α_i and $K>0$ Eq. (13) defines E_{mca} for each crystallite. We obtain the α_i by using $R(\Theta,\Phi)R(\xi)$ [cf. Eqs. (14) and (15)] to rotate $\hat{x}$, $\hat{y}$, and $\hat{z}$ into an orthonormal set of vectors corresponding to each easy direction $\hat{n}$ and exploit the identity employed in Eq. (16):

$$\alpha_1 \equiv \hat{M}\cdot R(\Theta,\Phi)R(\xi)\hat{x} = R(\theta,\phi)\hat{z}\cdot R(\Theta,\Phi)R(\xi)\hat{x}$$
$$= \hat{z}\cdot R^{-1}(\theta,\phi)R(\Theta,\Phi)R(\xi)\hat{x} \quad (29a)$$

and, similarly,

$$\alpha_2 \equiv \hat{M}\cdot R(\Theta,\Phi)R(\xi)\hat{y} = \hat{z}\cdot R^{-1}(\theta,\phi)R(\Theta,\Phi)R(\xi)\hat{y}, \quad (29b)$$

$$\alpha_3 \equiv \hat{M}\cdot R(\Theta,\Phi)R(\xi)\hat{z} = \hat{z}\cdot R^{-1}(\theta,\phi)R(\Theta,\Phi)R(\xi)\hat{z}. \quad (29c)$$

With these expressions we have

$$E_{\text{mca}} = -\frac{1}{2}K(\alpha_1^4+\alpha_2^4+\alpha_3^4) = -\frac{1}{2}K(a^4+b^4+c^4), \quad (30)$$

where a, b, and c remain defined by Eqs. (17b)–(17d). Substituting in Eq. (6) and then averaging over $0\leq\xi\leq2\pi$ yields[9]

$$\beta_{\text{rem}}^{100} = \frac{1}{16}\langle -99c^8+184c^6-102c^4+16c^2+1\rangle_{100} \quad (31)$$

with $c\equiv c(\rho,\sigma)$ specified by Eq. (19). This in turn leads to

$$\beta_{\text{rem}}^{100}(\rho) = \frac{1}{16}\Bigg\{\sin^8\rho\left[-\frac{637065}{128}\langle\cos^8\Theta\rangle+\frac{297297}{32}\langle\cos^6\Theta\rangle-\frac{343035}{64}\langle\cos^4\Theta\rangle+\frac{31185}{32}\langle\cos^2\Theta\rangle-\frac{3465}{128}\right]$$
$$+\sin^6\rho\left[\frac{42471}{4}\langle\cos^8\Theta\rangle-\frac{79233}{4}\langle\cos^6\Theta\rangle+\frac{45675}{4}\langle\cos^4\Theta\rangle-\frac{8295}{4}\langle\cos^2\Theta\rangle+\frac{115}{2}\right]$$
$$+\sin^4\rho\left[-\frac{29403}{4}\langle\cos^8\Theta\rangle+\frac{27405}{2}\langle\cos^6\Theta\rangle-7875\langle\cos^4\Theta\rangle+\frac{2835}{2}\langle\cos^2\Theta\rangle-\frac{153}{4}\right]$$
$$+\sin^2\rho[1782\langle\cos^8\Theta\rangle-3318\langle\cos^6\Theta\rangle+1890\langle\cos^4\Theta\rangle-330\langle\cos^2\Theta\rangle+8]-99\langle\cos^8\Theta\rangle+184\langle\cos^6\Theta\rangle$$
$$-102\langle\cos^4\Theta\rangle+16\langle\cos^2\Theta\rangle+1\Bigg\}, \quad (32)$$

in which the averages over $\cos^n\Theta$ are independent of sextant orientation and the 100 subscripts have been omitted. Either Eq. (26) or Eqs. (27) give

$$\langle\cos^n\Theta\rangle_{100}=\langle\cos^n\Theta\rangle =\frac{3}{n+1}\left[1-\frac{4}{\pi}\int_0^{\pi/4}d\Phi\left(\frac{\cos\Phi}{\sqrt{1+\cos^2\Phi}}\right)^{n+1}\right], \tag{33}$$

from which we obtain, again with the assistance of Ref. 7,

$$\langle\cos^2\Theta\rangle_{100}=\frac{1}{3}+\frac{2}{\pi\sqrt{3}}, \tag{34a}$$

$$\langle\cos^4\Theta\rangle_{100}=\frac{1}{5}+\frac{26}{15\pi\sqrt{3}}, \tag{34b}$$

$$\langle\cos^6\Theta\rangle_{100}=\frac{1}{7}+\frac{148}{105\pi\sqrt{3}}, \tag{34c}$$

$$\langle\cos^8\Theta\rangle_{100}=\frac{1}{9}+\frac{656}{567\pi\sqrt{3}}. \tag{34d}$$

Inserting these into Eq. (32), we thus find that the coefficient β in the law of approach to saturation from the remanent state of a {100}-easy polycrystal is

$$\beta_{\rm rem}^{100}(\rho)=\frac{8}{105}+\frac{1}{\pi\sqrt{3}}\left(\frac{1}{1260}+\frac{2}{35}\sin^2\rho-\frac{39}{112}\sin^4\rho+\frac{241}{420}\sin^6\rho-\frac{5093}{17920}\sin^8\rho\right). \tag{35}$$

Similar to the {111} case, averaging the right side of Eq. (31) over the unit sphere generates Eq. (32) with $\langle\cos^n\Theta\rangle_{100}$ replaced by $\langle\cos^n\Theta\rangle_{\rm unit\ sphere}$ from Eq. (24). This again yields $8/105=\beta_{\rm iso}$, so that we consistently recover the isotropic result.

IV. UNIAXIAL ANISOTROPY

For this comparatively simple case the moments in the remanent state are distributed uniformly over a hemisphere centered around $\hat{z}$, and the average of $f(\Theta,\Phi)$ over that region is

$$\langle f(\Theta,\Phi)\rangle_u=\frac{1}{2\pi}\int_0^{\pi/2}d\Theta\,\sin\Theta\int_0^{2\pi}d\Phi\,f(\Theta,\Phi), \tag{36}$$

the subscript u denoting uniaxial anisotropy; averaging over ζ is irrelevant since the hemisphere is invariant under rotation about $\hat{z}$. The normalized distribution of moments around $\hat{z}$ is clearly

$$g_u(\Theta)=\begin{cases}\dfrac{1}{2\pi}, & 0\leqslant\Theta\leqslant\dfrac{\pi}{2}\\ 0, & \dfrac{\pi}{2}<\Theta\leqslant\pi.\end{cases} \tag{37}$$

The magnetocrystalline anisotropy energy of a crystallite is

$$E_{\rm mca}=-K(\hat{n}\cdot\hat{M})^2=-K[\cos\theta\cos\Theta+\sin\theta\sin\Theta\cos(\phi-\Phi)]^2 \tag{38}$$

with $K>0$. Use of this in Eq. (6) yields

$$\beta_{\rm rem}^{u}=2\left\{\left[-1+5\sin^2\rho-\frac{35}{8}\sin^4\rho\right]\langle\cos^4\Theta\rangle_u+\left[1-\frac{9}{2}\sin^2\rho+\frac{15}{4}\sin^4\rho\right]\langle\cos^2\Theta\rangle_u+\frac{1}{2}\sin^2\rho-\frac{3}{8}\sin^4\rho\right\}=\frac{4}{15}, \tag{39}$$

since

$$\langle\cos^n\Theta\rangle_u=\frac{1}{n+1} \tag{40}$$

from Eq. (36). Therefore, $\beta_{\rm rem}^{u}$ for the remanent uniaxial state is independent of field direction and is equal to the well-known[1] result 4/15 for the isotropic state since $\langle\cos^n\Theta\rangle_u=\langle\cos^n\Theta\rangle_{\rm unit\ sphere}$, as a comparison of Eq. (40) and Eq. (24) shows.

V. REMARKS

To our knowledge, the distributions $g_{111}(\Theta)$ [Eq. (10c)] and $g_{100}(\Theta)$ [Eq. (27b)] of the moments in the remanent cubic states have not been derived previously by other authors. These distributions are significant milestones on the way to obtaining our principal results, the coefficients $\beta_{\rm rem}^{111}(\rho)$ and $\beta_{\rm rem}^{100}(\rho)$.

For the cubic systems our principal findings [Eq. (23) for $\beta_{\rm rem}^{111}(\rho)$ and Eq. (35) for $\beta_{\rm rem}^{100}(\rho)$] show that the law of approach to saturation depends on the initial *moment* distribution even if the underlying *crystallite* distribution is isotropic. If the initial *moment* distribution is isotropic, then $\beta=\beta_{\rm iso}=8/105$, as is well known and as we have demonstrated in checks of the results in this paper. If the initial moment distribution is that of the remanent state, however, then β depends on the easy-axis type ({111} or {100} easy) as well as on the angle ρ of the second saturating field with respect to the primary saturating field which created the remanent state. Put another way, β depends on magnetic history.

The fact that $\beta_{\rm rem}^{111}$ and $\beta_{\rm rem}^{100}$ are functions of ρ may appear surprising, but the following observation may serve to motivate it. For a single moment in a cubic system β is given by

$$\beta_{\rm cubic}^{\rm single}(\rho,\sigma)=2[\alpha_1^6+\alpha_2^6+\alpha_3^6-(\alpha_1^8+\alpha_2^8+\alpha_3^8)-2(\alpha_1^4\alpha_2^4+\alpha_2^4\alpha_3^4+\alpha_3^4\alpha_1^4)], \tag{41}$$

where $\alpha_1=\sin\rho\cos\sigma$, $\alpha_2=\sin\rho\sin\sigma$, $\alpha_3=\cos\rho$. Equation (41) follows directly from Eqs. (6) and (13), and it can also be found in Ref. 11; it shows that β for a single moment does depend on the field direction (and, therefore, on magnetic history). Hence, if we consider either of the remanent cubic states as a nonuniform collection of single moments, each having a field-dependent β and whose spatial distribution is highly directional, it is more comprehensible that β for the collection [Eq. (23) or Eq. (35)] also depends on the field direction.

We note that β for a single uniaxial moment also depends on the field direction; evaluation of Eq. (6) yields[12]

$$\beta_u^{\text{single}}(\rho) = \frac{1}{2}\sin^2 2\rho. \tag{42}$$

[On the basis of Eq. (41) and Eq. (42) we infer that β will depend on the field direction for a single moment of any anisotropy type.] The higher symmetry of the uniaxial case, however, produces a less directional distribution of moments whose β reproduces β_{iso}. Indeed, of the three cases we have considered, $\beta_{\text{rem}} = \beta_{\text{iso}}$ only for the uniaxial system, which is characterized by the lowest number (two) of equivalent easy directions. Our results suggest the general observation that $\beta_{\text{rem}} \neq \beta_{\text{iso}}$ for any other system since the number of equivalent easy directions will be greater than 2. A corollary of this observation is that, starting from the isotropic state, only the uniaxial system generates the moment distribution of the remanent state as it passes to saturation. In the uniaxial case each moment initially in the lower half-sphere of the isotropic state can simply flip, at no energy expense, to the equivalent easy direction in the upper half-sphere, thus forming the remanent distribution and implying $\beta_{\text{iso}} = \beta_{\text{rem}}$; the inference is that the remanent state cannot be formed by such flips in a system having more than two equivalent easy directions.

Experimental verification of Eq. (23) and Eq. (35) for the cubic anisotropy cases may well be possible; we suggest two approaches. First, if the second saturating field is applied in the same direction as the primary field which established the remanent state ($\rho = 0$), Eq. (23) predicts that $\beta_{\text{rem}}^{111}(0) = 0.074\,19$, 2.6% smaller than $\beta_{\text{iso}} = 8/105 = 0.076\,19$. Equation (35) yields $\beta_{\text{rem}}^{100}(0) = 0.076\,34$, 0.19% larger than β_{iso}. These differences are small but may be detectable, especially for a {111}-easy material. In real systems $M(H \to \infty)$ can contain a $1/H$ term arising from dislocation stresses, nonmagnetic impurities, and voids[1] which will make determination of β more difficult. If these mechanisms are isotropic, however, as seems likely, then the effects we have described might be more readily discernible by the second approach, measurement of the angular dependence. Figure 4 displays $\beta_{\text{rem}}^{111}(\rho)$ and $\beta_{\text{rem}}^{100}(\rho)$ as functions of the angle ρ between the first and second saturating fields. In each case the variation with ρ is quite apparent; the maximum peak-to-peak excursion is 7.3% of β_{iso} for $\beta_{\text{rem}}^{111}(\rho)$ and 1.8% of β_{iso} for $\beta_{\text{rem}}^{100}(\rho)$, both much larger than the corresponding differences for $\rho = 0$. Candidate systems for such investigation are melt-spun materials which can feature isotropically distributed single-domain crystallites.

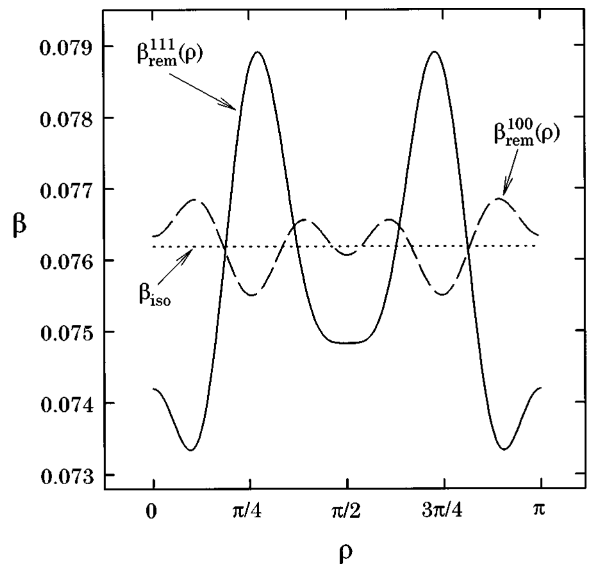

FIG. 4. $\beta_{\text{rem}}^{111}(\rho)$ (solid line) and $\beta_{\text{rem}}^{100}(\rho)$ (dashed line) vs the angle ρ between the primary and secondary saturating fields. The dotted line represents $\beta_{\text{iso}} = 8/105$ for the isotropic moment distribution.

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[1] S. Chikazumi, *Physics of Magnetism* (Wiley, New York, 1964), pp. 277, 280, 520.

[2] R. Becker and W. Döring, *Ferromagnetismus* (Springer, Berlin, 1939), p. 171.

[3] The relation between Θ and Φ on each great circle segment in Fig. 1 can be obtained from the condition $\hat{n} \cdot \hat{d}_i \times \hat{d}_j = 0$, where $\hat{n}$ is specified by Eq. (3) and $\hat{d}_i$, $\hat{d}_j$ are its endpoints.

[4] We use the $[0,2\pi]$ interval for convenience. Since the octant is threefold symmetric about $\hat{z}$, averaging over $0 \leq \zeta \leq 2\pi$ is equivalent to averaging over $\eta \leq \zeta \leq \eta + 2\pi/3$ with η an arbitrary fixed angle. The same consideration applies to the average over ξ leading to Eq. (18).

[5] S. Chikazumi, *Physics of Magnetism* (Ref. 1), p. 251.

[6] L. D. Landau, E. M. Lifshitz, and L. P. Pitaevskii, *Electrodynamics of Continuous Media*, Vol. 8 of Course of Theoretical Physics, 2nd edition (Pergamon, Oxford, 1984), p. 140.

[7] I. S. Gradshteyn and I. W. Ryzhik, *Table of Integrals, Series, and Products* (Academic, New York, 1965).

[8] We note that Eq. (23) can also be derived by using Eq. (8) for a single octant but averaging the integrand over σ; rotating the field about $\hat{z}$ is clearly equivalent to counterrotating the octant about $\hat{z}$. This alternate approach, however, does not yield the distribution $g_{111}(\Theta)$ explicitly.

[9] Again, we use the $[0,2\pi]$ interval for mathematical convenience. Since the sextant is fourfold symmetric about $\hat{z}$, averaging over $0 \leq \zeta, \xi \leq 2\pi$ is equivalent to averaging over $\eta \leq \zeta, \xi \leq \eta + \pi/2$ with η an arbitrary fixed angle.

[10] S. Chikazumi, *Physics of Magnetism* (Ref. 1), p. 250.

[11] S. Chikazumi, *Physics of Magnetism* (Ref. 1), p. 276.

[12] Equation (42) also follows from Eq. (39) evaluated with $\Theta = 0$, $\langle \cos^n \Theta \rangle_u = 1$; its implications that $\beta_u^{\text{single}}(0) = \beta_u^{\text{single}}(\pi/2) = 0$ and $\beta_u^{\text{single}}(0 < \rho < \pi/2) \neq 0$ are in agreement with classic findings of Stoner and Wohlfarth (Fig. 6 of Ref. 13) for an ellipsoidal ferromagnetic particle.

[13] E. C. Stoner and E. P. Wohlfarth, Philos. Trans. R. Soc. London, Ser. A **240**, 599 (1948).