

the Anderson treatment² of the magnetic impurity problem: $\tau^{-1}(E) = [\Delta/10N(0)]\rho_d(E)$. An effective relaxation rate can be defined as an average over the allowed transitions of the sum of the electron and hole scattering rates,²⁰

$$\tau^{-1}(\omega) = \omega^{-1} \int_{-\omega}^0 dE [\tau^{-1}(E) + \tau^{-1}(E + \omega)]. \quad (2)$$

Using a Lorentzian for $\rho_d(E)$, this effective relaxation rate gives rise to essentially the same result for ΔA as that obtained from the Kj ollerstr om calculation.

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Nonanalyticity of the Magnetization Curve: Application to the Measurement of Anisotropy in Polycrystalline Samples

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We studied the second derivative of the magnetization with respect to the magnetic field, d^2M/dH^2 , for polycrystalline materials having uniaxial anisotropy. Evidence is given of a singular point located at $H = H_A$, the anisotropy field. The observed sharp peak is associated with the infinity at $H = H_A$ of the d^2M/dH^2 curve for the single crystal in the hard direction. The detection of singularities is proposed as a new, and sometimes unique, method of measuring magnetic anisotropy using polycrystalline samples.

If we look at the reversible part of the magnetization curve $M(H)$ of a polycrystalline sample, we see that it is somewhat smooth, and we do not expect, in general, any kinds of sharp features. This is true if we limit ourselves to the observation of the magnetization, but we cannot be sure there will be the same behavior for the successive derivatives of M with respect to H , i.e., dM/dH , d^2M/dH^2 , etc. By carrying out measurements on a sintered sample of $\text{BaFe}_{12}\text{O}_{19}$ we indeed observed a sharp peak in d^2M/dH^2 versus magnetic field H , which is very similar to a cusp

located exactly at $H = -H_A = 2(K_1 + 2K_2)/M_s$ (the anisotropy field). This result is in agreement with the curves we have obtained starting from the magnetization curve of a polycrystalline specimen of uniaxial materials computed by Stoner and Wohlfarth,¹ and merely approximating the derivatives with the incremental ratios (Fig. 1). The most interesting features of this phenomenon are its sharpness and the coincidence with the anisotropy field.

A mathematical approach starting from the usual phenomenological treatment of magnetic

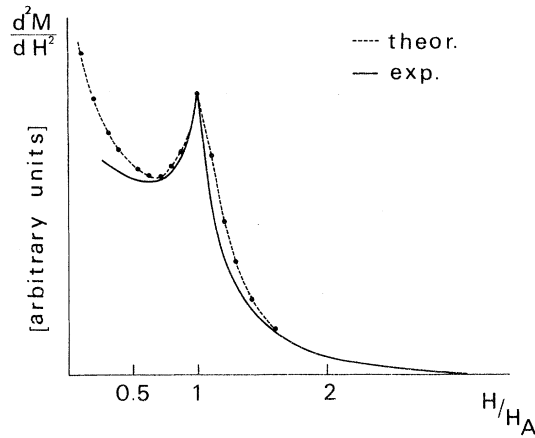


FIG. 1. Experimental and computed curves for the dependence of d^2M/dH^2 on the magnetic field H . The theoretical curve is obtained from the data of Stoner and Wohlfarth (Ref. 1).

anisotropy may lead to a good theoretical interpretation based on the analysis of the mathematical singularities of the function $M(H)$.

As is well known,² in the case of a uniaxial crystal the magnetocrystalline anisotropy energy is expressed to a first approximation by the formula

$$E_K = K_1 \sin^2\theta + K_2 \sin^4\theta,$$

where θ denotes the angle between the symmetry axis and the magnetization [Fig. 2(a)].

The magnetization curve $M(H)$ of a single crystal magnetized in the hard direction, that is, with the magnetic field forming an angle $\varphi = \pi/2$ with the crystal axis, is characterized by an angular point exactly at $H = H_A$. For $\varphi \neq \pi/2$ the curve $M(H)$ is regular but the curvature for $H = H_A$ increases indefinitely as φ approaches $\pi/2$.

Therefore in studying the singular behavior of the magnetization curve of a polycrystalline aggregate it is not necessary to average over all possible orientations ($-\pi/2 \leq \varphi \leq +\pi/2$), but an examination can be limited to a small interval centered around the point $\varphi = \pi/2$ and $H = H_A$. The

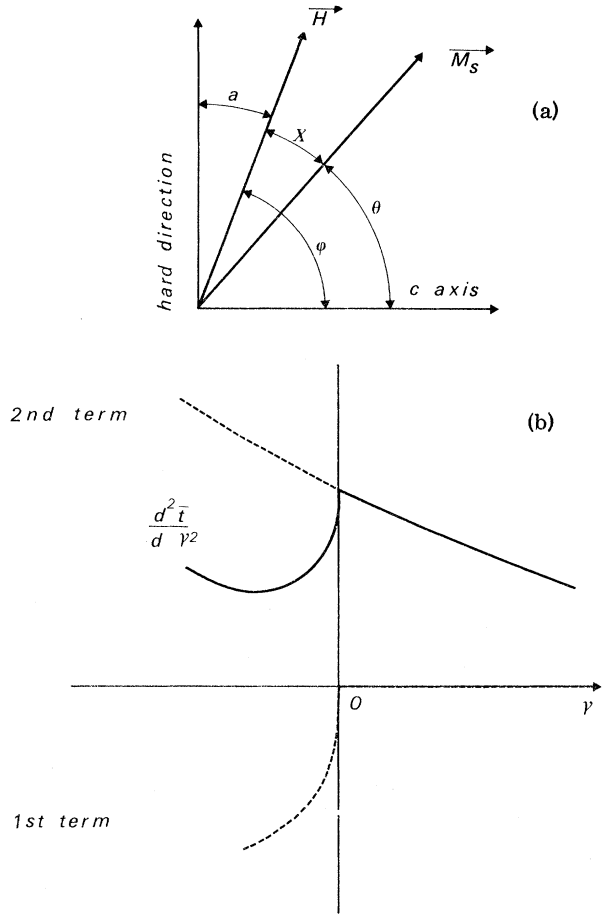


FIG. 2. (a) Coordinates used for the magnetic field \vec{H} and the magnetization \vec{M}_s in the crystallites. (b) Behavior of the second derivative of the mean reduced magnetization \bar{t} with respect to the reduced field γ , $d^2\bar{t}(\gamma)/d\gamma^2$. The averaging is performed in the vicinity of the singular point $\gamma = 0$ ($H = |H_A|$).

total energy can be written

$$E_t = E_h + E_H = K_1 \sin^2\theta + K_2 \sin^4\theta - M_s H \cos(\theta - \varphi),$$

where E_H is the magnetostatic energy. By defining the infinitesimal quantities $x = \theta - \varphi$, $a = -\varphi + \pi/2$, and $\gamma = (H - |H_A|)/|H_A|$, disregarding the terms of order higher than 4, we have

$$E_t = K_1[1 - (x-a)^2 + (x-a)^4/3] + K_2[1 - 2(x-a)^2 + 5(x-a)^4/3] + M_s H_A(\gamma + 1)(1 - x^2/2 + x^4/24).$$

The equilibrium condition $\partial E_t / \partial x = 0$ gives

$$Lx^3 + x + a = 0, \tag{1}$$

where higher-order terms have been neglected and L is $L = (K_1 + 6K_2)/(2K_1 + 4K_2)$. In addition we define the "reduced magnetization" as the dimensionless quantity $t = (M_s - M)/M_s = 1 - \cos x \cong x^2/2$, that is, the magnetization measured, starting from saturation, in units of M_s . For polycrystalline samples with

individual crystallites at random orientation we have

$$\bar{t}(\gamma) = 2 \int_{a_0}^0 t(\gamma, a) da \cong 2 \int_{a_0}^0 (x^2/2) da, \quad (2)$$

where a_0 is the maximum a limiting the small interval under examination. By taking the derivative with respect to γ we obtain

$$d\bar{t}(\gamma)/d\gamma = 2 \int_{a_0}^0 x(\partial x/\partial \gamma) da, \quad (3)$$

but $\partial x/\partial \gamma = -x \partial x/\partial a$, as can be easily demonstrated by differentiating Eq. (1). So, Eq. (3) becomes

$$d\bar{t}(\gamma)/d\gamma = -2 \int_{x_0}^{x_1} x^2 dx = -\frac{2}{3}(x_1^3 - x_0^3), \quad (4)$$

where

$$x_0 = x(\gamma, a_0) = \left[\frac{a_0}{2L} + \left(\frac{a_0^2}{4L^2} + \frac{\gamma^3}{27L^3} \right)^{1/2} \right]^{1/3} + \left[\frac{a_0}{2L} - \left(\frac{a_0^2}{4L^2} + \frac{\gamma^3}{27L^3} \right)^{1/2} \right]^{1/3},$$

$$x_1 \equiv [(-\gamma^+|\gamma|)/2L]^{1/2} = \begin{cases} (-\gamma/L)^{1/2} & \text{for } \gamma < 0, \\ 0 & \text{for } \gamma > 0; \end{cases}$$

the upper limit in the integral is a function with a singular point at $\gamma=0$. It is worth noting that this fact is a direct consequence of the existence of an angular point in the function $t(\gamma, a=0)$ (single crystal in the hard direction). The second derivative is

$$\frac{d^2\bar{t}(\gamma)}{d\gamma^2} = L^{-1} \left(\frac{-\gamma^+|\gamma|}{2L} \right)^{1/2} + 2x_0^2 \frac{\partial x_0}{\partial \gamma}. \quad (5)$$

The two terms have the behavior shown in Fig. 2(b). The sum of the two terms gives rise to a curve with a cusp on the left side of $\gamma=0$; this is very similar in shape to the singularity we have observed in our experimental and theoretical curves.

The phenomenon described is probably not peculiar to uniaxial materials but it may be more general and its origin related to the properties of polycrystalline aggregates of anisotropic magnetic materials. For this we are trying to extend the mathematical approach to the other anisotropy symmetries and verify experimentally the predictions that seem to be similar to the case explained here.

The detection of singularities in the magnetization curve can be considered a new method to measure the anisotropy field in polycrystalline samples. Such a technique is advantageous because it is not necessary that the grains be single domain since the measurements are performed in the descending part of the magnetization curve where the only magnetization proces-

ses are reversible rotations and the domain walls do not play any role. Moreover, the results obtained with this method are insensitive to the distribution in size and orientation of the crystallite in the sample: On the contrary, accurate amplitude analysis of the peaks could, in principle, reveal the texture of the specimen.

A case in which this method is particularly suitable is that of materials with noncollinear spin order: It allows one to separate the anisotropy from the exchange contribution to the differential susceptibility at high fields.

We have carried out measurements by the pulsed-magnetic-field technique on the two series of hexagonal compounds, $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{19}$ and $\text{SrFe}_{12-x}\text{Ga}_x\text{O}_{19}$; and we were able to measure the anisotropy field with a precision of 2% in polycrystalline sample up to values as high as 70 000 Oe; this allowed us to demonstrate that some of these compounds have a noncollinear spin order. These results will be reported extensively elsewhere.³

The model discussed here is a good approximation for hard magnetic materials, but as H_A becomes smaller, some accounting for interparticle interaction may be needed.

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