Phase Transitions in a Uniaxial Ferromagnet

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(Received 23 June 1969)

The phase transitions occurring in a uniaxial ferromagnet are studied for all temperatures up to the Curie temperature. The stability limits are obtained in the molecular-field approximation, and the critical dynamic behavior is discussed for a simple model taking spin-lattice relaxation effects into account. For fields applied in the easy direction and temperatures below some characteristic temperature $T_1<T_C$, and for fields applied in the hard direction and all temperatures $T<T_C$, the instability occurs with respect to angular deviations of the magnetization from its equilibrium direction, the associated soft mode is a ferromagnetic resonance (or perpendicular relaxation) mode, and one obtains critical fluctuations of magnetization direction. The same holds approximately true for fields applied at an arbitrary angle and temperatures $T<T_1$. For fields applied in the easy direction and temperatures in the interval $T_1<T<T_C$, the instability occurs with respect to deviations of magnetization magnitude, the associated soft mode is a parallel relaxation mode, and one obtains critical fluctuations of magnetization magnitude. The type of singularity of the static susceptibility and the behavior of the soft-mode frequencies at the stability limits are given for the different cases.

I. INTRODUCTION

PHASE transitions occurring in anisotropic ferromagnets as a function of the applied field at temperatures small compared to the Curie temperature are well understood in principle. In this temperature region, it is a good approximation to consider the magnitude M of the magnetization vector $\mathbf{M}$ as constant, such that the magnetic state is characterized by the magnetization direction alone.

For simplicity, we shall disregard the long-range effects of the dipolar interactions between the magnetic moments, and take only states of uniform magnetization into account, both for thermodynamic equilibrium and for deviations from equilibrium. Usually, these magnetostatic effects require special attention. Thus, in samples of arbitrary shape in uniform external field, the magnetization direction at thermal equilibrium is nonuniform. Even in ellipsoidal samples, instabilities occur in general with respect to nonuniform deviations from the uniform equilibrium state, e.g., with respect to curling or buckling modes in the case of an infinite cylinder (see Ref. 1 for a discussion of these problems and further references). Therefore, the discussion of the present paper applies strictly only to the case of a thin film with the easy direction in the film plane, for which the instabilities do occur with respect to uniform rotation. However, in very hard magnetic materials, magnetostatic effects are less important, and the properties will be approximately shape-independent. It will turn out that our results are especially significant for such hard magnetic materials, which may serve as partial justification of the simplifying assumption. We further assume an ideal material and neglect local nucleation of magnetization reversal at imperfections, etc., which would reduce the stability limits.

We consider a ferromagnet with uniaxial anisotropy energy of second order with an easy axis along the z direction, for applied fields in the x-z plane. At low temperatures, the behavior of this system is well known.[2] It has a first-order phase transition[3] with the thermodynamic phase boundary along the line segment

$$-H_K<H_x<+H_K, \quad H_z=0, \tag{1}$$

where H_K is the anisotropy field. At both sides of the phase boundary (1) there exist regions of metastability; the stability limits are given by the critical curve[2]

$$H_x^{2/3}+H_z^{2/3}=H_K^{2/3}, \tag{2}$$

which is shown in Fig. 1. It is these stability limits rather than the thermodynamic phase boundary which are of dynamical significance.

It is useful to visualize the H_x-H_z plane as a topological surface which has a cut between the points $H_x=\pm H_K$, $H_z=0$, and which inside of the critical curve consists of two sheets, corresponding to the two phases $M_z>0$ and $M_z<0$, respectively. The stability limits determine the edges of the two sheets: The $M_z>0$ sheet has its edge at the lower section, the $M_z<0$ sheet at the upper section of the critical curve.

The points $H_x=\pm H_K$, $H_z=0$ are critical points of the phase transition: One may pass from a state $M_z>0$ to a state $M_z<0$ either discontinuously by crossing the edge of the $M_z>0$ sheet and changing to the other, or continuously along a path bypassing the critical points. For a path passing through the critical points, and especially for fields in the hard direction, the magnetization is continuous, i.e., the phase transition is of second order.

Since the magnitude of the magnetization is taken as constant, the instability occurs with respect to angular deviations of the magnetization. The susceptibility

[1] W. F. Brown, Jr., *Micromagnetics* (Wiley-Interscience, Inc., New York, 1963).

[2] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press Ltd., London, 1960), Sec. 37.

[3] This is a special case of a first-order phase transition, because only one of the first derivatives, $\partial F/\partial H_z=-M_z$, of the Gibbs free energy $F(\mathbf{H},T)$ has a discontinuity. The entropy $S=-\partial F/\partial T$ is continuous at the thermodynamic phase boundary, i.e., there is no latent heat.

tensor has principal axes parallel and perpendicular to $\mathbf{M}$, and it is the perpendicular susceptibility $\chi_\perp$ (corresponding to the axis perpendicular to $\mathbf{M}$ in the x-z plane) which becomes singular at the stability limit; the parallel susceptibility $\chi_\parallel$ is zero in the approximation $M=\text{const}$. Consequently, the soft dynamical mode, the frequency of which becomes zero at the stability limit, is the ferromagnetic resonance mode. Such soft-mode behavior of ferromagnetic resonance is in fact observed experimentally in thin magnetic films.[4,5] If magnetostatic interactions are taken into account, one finds that instabilities with respect to nonuniform deviations from equilibrium are associated with a softening of the corresponding nonuniform magnetostatic spin-wave modes (see Chap. 7 of Ref. 1).

In the following, we drop the assumption $M=\text{const}$, and study the behavior of a uniaxial ferromagnet at all temperatures in the molecular field approximation. In Sec. II, we derive an expression for the free energy of the system. The stability limits are obtained in Sec. III from the second variation of the free energy. Of particular interest is the result that for fields in the easy direction and temperatures close to the Curie temperature, the instability occurs with respect to a parallel mode.[6] In Sec. IV, we discuss the dynamic behavior at the stability limits for a system with spin-lattice relaxation with the help of a modified Bloch equation.

II. FREE ENERGY

The behavior of the system at given temperature and field is determined by the density matrix ρ which minimizes the free energy

$$\mathcal{F}=\mathcal{E}-T\mathcal{S}, \tag{3}$$

where

$$\mathcal{E}=\langle\mathcal{H}\rangle=\mathrm{tr}(\rho\mathcal{H}) \tag{4}$$

is the energy, and

$$\mathcal{S}=-k_B\langle\ln\rho\rangle=-k_B\,\mathrm{tr}(\rho\ln\rho) \tag{5}$$

is the entropy of the system.

We consider a uniaxial ferromagnet consisting of N equivalent spins $\mathbf{S}_i$, which is described by a Hamiltonian of the form

$$\mathcal{H}=\mathcal{H}^{\mathrm{exch}}+\mathcal{H}^{\mathrm{anis}}+\mathcal{H}^{\mathrm{Zeeman}}. \tag{6}$$

The first term is the isotropic exchange energy

$$\mathcal{H}^{\mathrm{exch}}=-\tfrac{1}{2}\sum_{ij}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j. \tag{7}$$

The anisotropy energy may consist of a combination of anisotropic exchange and crystal-field-type anisotropy:

$$\mathcal{H}^{\mathrm{anis}}=-\tfrac{1}{2}\sum_{ij}K_{ij}S_i^zS_j^z -\tfrac{1}{2}\hat{L}\sum_i[(S_i^z)^2-\tfrac{1}{3}S(S+1)]. \tag{8}$$

The last term in Eq. (6) represents the Zeeman energy in the applied field $\mathbf{H}$:

$$\mathcal{H}^{\mathrm{Zeeman}}=-\mathbf{H}\cdot\sum\mathbf{S}_i, \tag{9}$$

where H is measured in units of $g\mu_B$.

We calculate the free energy in molecular-field approximation (MFA) for a trial density matrix of the form

$$\rho=\prod_i(Z_i)^{-1}\exp(\beta\mathbf{\Lambda}\cdot\mathbf{S}_i),\quad \beta=(k_BT)^{-1} \tag{10}$$

with $\mathbf{\Lambda}$ as a variational parameter. We take the same value of $\mathbf{\Lambda}$ for all spins, because we are interested only in states of uniform magnetization. Then, the Z_i are all equal and are given by

$$Z_i=\mathrm{tr}_i\exp(\beta\mathbf{\Lambda}\cdot\mathbf{S}_i)=Z_S(\beta S\Lambda), \tag{11}$$

where

$$Z_S(x)=\sinh\left(\frac{2S+1}{2S}x\right)\Big/\sinh\frac{x}{2S}. \tag{12}$$

With the density matrix (10), the average spin moment $\langle\mathbf{S}\rangle$ can be calculated as a function of $\mathbf{\Lambda}$. For the average magnetization $\boldsymbol{\sigma}$ defined by

$$\langle\mathbf{S}\rangle=S\boldsymbol{\sigma}, \tag{13}$$

one obtains

$$\boldsymbol{\sigma}=(\mathbf{\Lambda}/\Lambda)B_S(\beta S\Lambda), \tag{14}$$

where

$$B_S(x)=\frac{1}{Z_S}\frac{dZ_S}{dx}=\frac{2S+1}{2S}\coth\frac{2S+1}{2S}x-\frac{1}{2S}\coth\frac{x}{2S} \tag{15}$$

is the Brillouin function. The free energy (3) is most

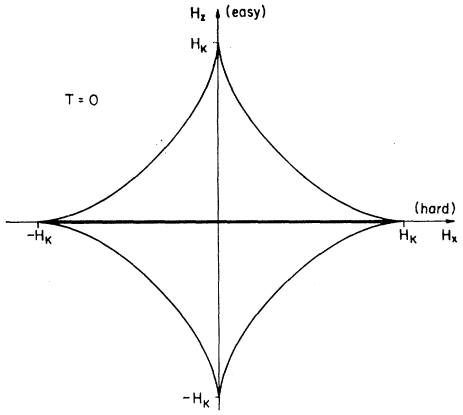

FIG. 1. Critical curve for $T=0$.

[4] R. H. Kingston and P. E. Tannenwald, J. Appl. Phys. **29**, 232 (1958).

[5] P. Wolf, Z. Physik **160**, 310 (1960).

[6] These results were presented at the Meeting of the Arbeitsgemeinschaft Magnetismus, Amsterdam, 1969 (unpublished).

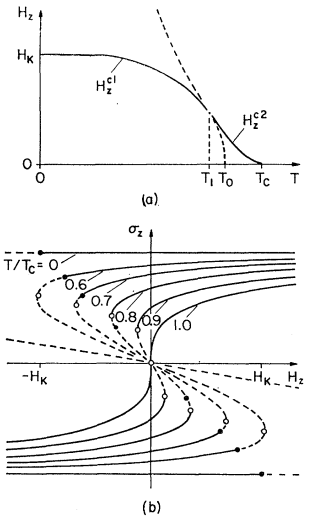

FIG. 2. Magnetization reversal in the easy direction for the case $S=1, K/J=0.2$. (a) Stability limits in the (H_z,T) plane. (b) Magnetization curves for various temperatures with stability limits against perpendicular (●) and parallel (○) mode.

easily expressed in terms of $\boldsymbol{\sigma}$; we find

$$F\equiv \mathfrak{F}/N=-\tfrac{1}{2}S^2[J-\tfrac{1}{3}L(\sigma)]\sigma^2 -\tfrac{1}{2}S^2[K+L(\sigma)]\sigma_z^2-S\mathbf{H}\cdot\boldsymbol{\sigma}-k_BT\eta(\sigma). \quad (16)$$

Here, $k_B\eta(\sigma)$ is the single spin entropy in MFA. It is given in terms of the inverse Brillouin function $B_S^{-1}(\sigma)$ by

$$\eta(\sigma)=\ln Z_S\{B_S^{-1}(\sigma)\}-\sigma B_S^{-1}(\sigma), \quad (17)$$

whence

$$d\eta(\sigma)/d\sigma=-B_S^{-1}(\sigma)\equiv-\beta S\Lambda. \quad (18)$$

Further,

$$J=\sum_i J_{ij}, \quad K=\sum_i K_{ij} \quad (19)$$

are the constants of isotropic and anisotropic exchange, respectively, and $L(\sigma)$ is an effective, magnetization-dependent crystal-field anisotropy constant, which can be obtained from the results given by Wolf.[7] It is related to the function $p(\sigma)$ of Ref. 7 by

$$L(\sigma)=\hat{L}p(\sigma)/(S^2\sigma^2) \quad (20)$$

and varies between the limits

$$L(0)=\tfrac{3}{5}\hat{L}\left(1-\frac{1}{2S}\right)\frac{2S+3}{2S+2}, \qquad L(1)=\hat{L}(1-1/2S). \quad (21)$$

We assume $K+L(\sigma)>0$ for all σ, which guarantees that the z axis is an easy axis of magnetization at all temperatures.

Since we have eliminated the parameter $\boldsymbol{\Lambda}$ in favor of the relative magnetization $\boldsymbol{\sigma}$, we can now consider $\boldsymbol{\sigma}$ as variational parameter to be determined by minimizing the free energy (16). The equilibrium condition

$$\partial F/\partial\boldsymbol{\sigma}=0 \quad (22)$$

in connection with Eq. (18) shows that, at equilibrium, $\boldsymbol{\Lambda}$ is equal to the molecular field defined by

$$\mathbf{H}^{\mathrm{mol}}=-(1/NS)(\partial\langle\mathfrak{IC}\rangle/\partial\boldsymbol{\sigma}). \quad (23)$$

III. STABILITY LIMITS

A solution $\boldsymbol{\sigma}$ of Eq. (22) represents a stable equilibrium state if the free energy increases for all small deviations $\delta\boldsymbol{\sigma}$, i.e., if the quadratic form

$$\delta^2F=\tfrac{1}{2}\delta\boldsymbol{\sigma}\cdot\frac{\partial^2F}{\partial\boldsymbol{\sigma}\partial\boldsymbol{\sigma}}\cdot\delta\boldsymbol{\sigma} \quad (24)$$

is positive definite, or, in other words, if both eigenvalues of the matrix $\partial^2F/(\partial\boldsymbol{\sigma}\partial\boldsymbol{\sigma})$ are positive. The stability limits are thus determined by the condition that the smallest eigenvalue $\lambda_{\min}(\mathbf{H},T)$ becomes zero. Since the matrix of second derivatives of the free energy is related to the reciprocal static susceptibility tensor by

$$S^2\chi^{-1}=\partial^2F/\partial\boldsymbol{\sigma}\partial\boldsymbol{\sigma}, \quad (25)$$

this condition simply means that one principal value of the static susceptibility tensor becomes singular at the stability limit. The nature of the instability is determined by the eigenvector $\delta\boldsymbol{\sigma}$ belonging to $\lambda_{\min}$. If $\delta\boldsymbol{\sigma}\perp\boldsymbol{\sigma}$, the instability occurs with respect to angular variations (perpendicular or "rotational" mode); if $\delta\boldsymbol{\sigma}\parallel\boldsymbol{\sigma}$, it occurs with respect to deviations in magnitude (parallel or "shrinking" mode); in all other cases, it occurs with respect to a mixed mode.

Although the presence of the magnetization-dependent crystal-field anisotropy constant $L(\sigma)$ in the free-energy expression (16) causes no difficulties in principle, it does lead to lengthy expressions for the derivatives of the free energy. For simplicity, we therefore restrict the following discussion to the case where the anisotropy is produced by anisotropic exchange alone. The effects of the two types of anisotropy are expected to be qualitatively very similar, and we shall indicate in limiting cases ($T=0$ and $T=T_C$) how the results change when both types are present.

For $L=0$, the equilibrium condition (22) can be written in the form

$$S^2(J\sigma\mathbf{s}+K\sigma_z\boldsymbol{\zeta})+S\mathbf{H}=k_BTB_S^{-1}(\sigma)\mathbf{s}\equiv S\boldsymbol{\Lambda}, \quad (26)$$

where $\boldsymbol{\zeta}$ and $\mathbf{s}$ are unit vectors in the z direction and in the direction of $\boldsymbol{\sigma}$, respectively. For the tensor of second derivatives of the free energy, we obtain

$$(1/S^2)\partial^2F/\partial\boldsymbol{\sigma}\partial\boldsymbol{\sigma}\equiv\chi^{-1}=\chi_f^{-1}-(J\mathbf{1}+K\boldsymbol{\zeta}\boldsymbol{\zeta}). \quad (27)$$

Here, χ_f is the reciprocal static free-spin susceptibility tensor

$$\chi_f^{-1}=\chi_{f\parallel}^{-1}\mathbf{ss}+\chi_{f\perp}^{-1}(\mathbf{1}-\mathbf{ss}) \quad (28)$$

[7] W. P. Wolf, Phys. Rev. **108**, 1152 (1957).

with principal values

$$\chi_{f\parallel}^{-1}=\frac{k_BT}{S^2}\frac{dB_S^{-1}(\sigma)}{d\sigma}\equiv\frac{1}{S}\frac{d\Lambda}{d\sigma},$$
$$\chi_{f\perp}^{-1}=\frac{k_BT}{S^2}\frac{B_B^{-1}(\sigma)}{\sigma}\equiv\frac{1}{S}\frac{\Lambda}{\sigma}. \tag{29}$$

The Curie temperature is determined as the stability limit of the paramagnetic phase $\sigma=0$ in zero field. Because of

$$\chi_f^{-1}[3k_BT/S(S+1)]1 \quad \text{for } \sigma\to 0, \tag{30}$$

Eq. (27) becomes independent of **s**, and we find the eigenvalues

$$\chi_{zz}^{-1}=[3/S(S+1)]k_BT-(J+K),$$
$$\chi_{xx}^{-1}=\chi_{yy}^{-1}=[3/S(S+1)]k_BT-J. \tag{31}$$

The smallest eigenvalue is χ_{zz}^{-1}; it becomes zero for

$$k_BT_C=\tfrac{1}{3}S(S+1)(J+K). \tag{32}$$

At this temperature, the paramagnetic phase becomes unstable with respect to spontaneous magnetization in the z direction. In the presence of crystal-field anisotropy, Eq. (32) takes the form

$$k_BT_C=\tfrac{1}{3}S(S+1)[J+K+\tfrac{2}{3}L(0)]. \tag{33}$$

Next, we discuss the stability limits occurring for fields in the easy and the hard direction at temperatures below T_C. Figures 2 and 3 show the magnetization curves and the critical fields for these two cases.

For fields $\mathbf{H}=\{0,0,H_z\}$ in the easy direction, the state

$$\mathbf{s}=\boldsymbol{\zeta}, \quad S^2(J+K)\sigma+SH_z=k_BTB_S^{-1}(\sigma) \tag{34}$$

is stable for all $H_z>0$. We are interested in the stability limit of the metastable state

$$\mathbf{s}=-\boldsymbol{\zeta}, \quad S^2(J+K)\sigma-SH_z=k_BTB_S^{-1}(\sigma). \tag{35}$$

At low temperature, the two smallest eigenvalues are

$$\chi_{xx}^{-1}=\chi_{yy}^{-1}\equiv\chi_\perp^{-1}=\chi_{f\perp}^{-1}-J; \tag{36}$$

they become zero for a critical value $\sigma=\sigma_z^{c1}(T)$. At the corresponding critical field $H_z^{c1}(T)$ determined by Eq. (35), the state (35) becomes unstable with respect to a perpendicular mode, and a first-order phase transition occurs to the state (34). At $T=0$, this field is equal to the anisotropy field:

$$H_z^{c1}(0)=SK\equiv H_K. \tag{37}$$

Its temperature dependence is that of a Weiss-Brillouin curve which goes to zero at a temperature

$$k_BT_0=\tfrac{1}{3}S(S+1)J<k_BT_C, \tag{38}$$

as shown in Fig. 2. At temperatures close to the Curie temperature, the eigenvalue

$$\chi_{zz}^{-1}\equiv\chi_\parallel^{-1}=\chi_{f\parallel}^{-1}-(J+K), \tag{39}$$

which becomes zero for a critical value $\sigma=\sigma_z^{c2}(T)$, must determine the transition. The corresponding critical field $H_z^{c2}(T)$ determined by Eq. (35) is at $T=0$ equal to the sum of exchange and anisotropy field:

$$H_z^{c2}(0)=S(J+K)\equiv H_J+H_K. \tag{40}$$

It becomes equal to H_z^{c1} at a temperature T_1 somewhat below T_0 and goes to zero at the Curie temperature, as shown in Fig. 2. Therefore, in the temperature region $T_1<T<T_C$, the instability occurs with respect to the parallel mode.

For fields $\mathbf{H}=\{H_x,0,0\}$ in the hard direction, the instability occurs always with respect to the perpendicular mode. For sufficiently large values of H_x, the stable state is given by

$$\mathbf{s}=\boldsymbol{\xi}, \quad S^2J\sigma+SH_x=k_BTB_S^{-1}(\sigma). \tag{41}$$

The smallest eigenvalue

$$\chi_{zz}^{-1}=\chi_{f\perp}^{-1}-(J+K) \tag{42}$$

becomes zero for a value $\sigma=\sigma_x^c$ which determines the critical field $H_x^c(T)$ by Eq. (41). At $T=0$, this field is again equal to the anisotropy field:

$$H_x^c(0)=SK\equiv H_K, \tag{43}$$

and its temperature dependence is that of a Weiss-Brillouin curve going to zero at the Curie temperature, as shown in Fig. 3. At $H=H_x^c(T)$, there occurs a second-order phase transition, and for fields $H_x<H_x^c(T)$ the stable state is determined by

$$\sigma_x=H_x/H_K, \quad S^2(J+K)\sigma=k_BTB_S^{-1}(\sigma). \tag{44}$$

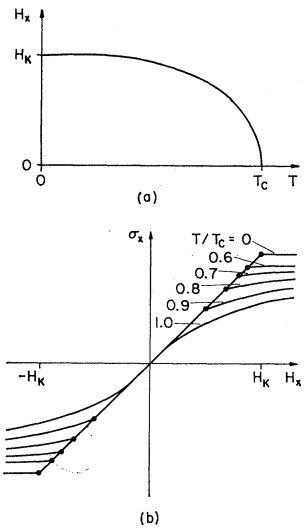

FIG. 3. Magnetization reversal in the hard direction for the case $S=1$, $K/J=0.2$. (a) Second-order phase boundaries in the H_x-T plane. (b) Magnetization curves for various temperatures.

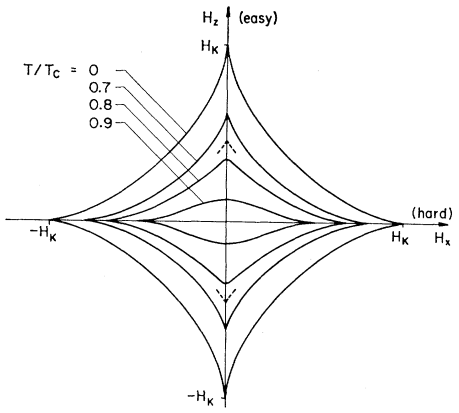

FIG. 4. Critical curves for various temperatures. $S=1$, $K/J=0.2$.

In this region, only the magnetization direction depends on the field; its magnitude is a function of temperature alone.

In the presence of crystal-field anisotropy, the zero-temperature anisotropy field in Eqs. (37), (40), and (43) is given by

$$H_K=S[K+L(1)]. \tag{45}$$

The type of singularity of the static susceptibility at the stability limits $H_z^{c1,2}$ and H_x^c is obtained easily by observing that the eigenvalues given in Eqs. (36), (39), and (42) go to zero like $\sigma-\sigma^c$. One finds for the three cases

$\mathbf{H}=\{0,0,H_z\}$:

$$\chi_\perp=\frac{\gamma_1(T)}{H_z^{c1}(T)-H_z}, \tag{46}$$

$$\chi_{\parallel}=\frac{\gamma_2(T)}{[H_z^{c2}(T)-H_z]^{1/2}}; \tag{47}$$

$\mathbf{H}=\{H_x,0,0\}$:

$$\chi_\perp=\frac{\alpha(T)}{H_x-H_x^c(T)}, \tag{48}$$

with temperature-dependent coefficients $\gamma_{1,2}(T)$ and $\alpha(T)$. As stated in the Introduction, the discussion in this paper is restricted to the spatially uniform case. We note in passing that for the static response to non-uniform fields with a small but finite wave number q, the denominators of Eqs. (46)–(48) increase by terms of order q^2.

Finally, we discuss the case of fields $\mathbf{H}=\{H_x,0,H_z\}$ applied at an arbitrary angle. The magnetization direction $\mathbf{s}$ is coplanar with the anisotropy axis $\boldsymbol{\zeta}$ and the field $\mathbf{H}$, as may be seen from Eq. (26), and can therefore be described by its angle with respect to the z axis:

$$\mathbf{s}=\{\sin\gamma, 0, \cos\gamma\}. \tag{49}$$

One eigenvalue is χ_{yy}^{-1}, corresponding to rotations out of the plane; however, the stability limit is always determined by one of the other two eigenvalues. The components of χ^{-1} in the x-z plane in a system of axes parallel and perpendicular to the magnetization direction $\mathbf{s}$ are found to be

$$\begin{aligned}(\chi^{-1})_{\parallel,\parallel}&=\chi_{f\parallel}^{-1}-(J+K\cos^2\gamma),\\ (\chi^{-1})_{\perp,\perp}&=\chi_{f\perp}^{-1}-(J+K\sin^2\gamma),\\ (\chi^{-1})_{\parallel,\perp}&=-K\sin\gamma\cos\gamma.\end{aligned} \tag{50}$$

The presence of the nondiagonal term $(\chi^{-1})_{\parallel,\perp}$ shows that, for $\gamma\neq 0, \frac{1}{2}\pi$, the instability occurs always with respect to a mixed mode. We have calculated numerically the set of critical values of (σ,γ) for which the smallest eigenvalue of (50) becomes zero. The critical curve is obtained by substituting this set into Eq. (26) and solving for $\mathbf{H}$. Figure 4 shows the critical curves for various temperatures for the case $S=1$, $K/J=0.2$.

For low temperatures, the mode is still approximately perpendicular, because $(\chi^{-1})_{\parallel,\parallel}$ becomes very large, and we recover the behavior discussed in the Introduction. In the temperature region $T_1<T<T_C$, on the other hand, we find a gradual transition from a perpendicular mode at $\mathbf{H}=\{H_x^c(T),0,0\}$ to a parallel mode at $\mathbf{H}=\{0,0,H_z^{c2}(T)\}$, and the critical curves no longer show a cusp in the easy direction. Close to the Curie temperature, we find for the critical curves the analytic expression

$$(H_x/H_x^c)^2+(H_z/H_z^{c2})^{2/3}=1. \tag{51}$$

We have calculated the stability limits by using the MFA. Because of the limited validity of this approximation, the quantitative results have to be considered with the usual reservation. It is, for instance, well known that for short-range interactions the Curie temperature is considerably smaller than the value obtained in Eq. (32). Consequently, the temperature dependence of the critical fields will be different from that shown in Figs. 2 and 3. We believe, nevertheless, that the qualitative picture is correct, and that especially for high temperatures the instability in the easy direction does occur with respect to a parallel mode.

Moreover, Shtrikman and Callen[8] have shown that relations between single spin properties, from which the molecular field parameter $\beta\Lambda$ has been eliminated, have a wider validity than the MFA itself. We therefore expect the form of the critical curves in Fig. 4 to be essentially correct. The same argument applies to the values of the critical fields $H_z^{c1,2}$ and H_x^c, if they are represented as functions of the zero-field magnetization σ_0 instead of the temperature. These functions are shown in Fig. 5.

IV. DYNAMIC BEHAVIOR

The results obtained suggest the following dynamic properties at the stability limits of the system. For

[8] H. B. Callen and S. Shtrikman, Solid State Commun. 3, 5 (1965).

fields applied in the easy direction at temperatures $T<T_1$ and for fields applied in the hard direction at all temperatures $T<T_C$, the soft dynamical mode associated with the transition is the ferromagnetic resonance (or perpendicular relaxation) mode, and one obtains critical fluctuations of magnetization direction. The same will hold approximately true for fields applied at an arbitrary angle at temperatures $T<T_1$. In the temperature interval $T_1<T<T_C$ for fields applied in the easy direction, on the other hand, the ferromagnetic resonance frequency remains finite at the stability limit H_z^{c2}, and we expect a soft parallel mode and critical fluctuations of magnetization magnitude.

For the Heisenberg Hamiltonian given in Eqs. (6)–(9) with the field applied in the z direction, the total spin component in the z direction is a constant of the motion, and the parallel mode will be a spin-diffusion mode with purely imaginary frequency

$$\Omega=-iDq^2 \tag{52}$$

for small wave numbers q. The diffusion constant D is related to the reciprocal parallel susceptibility by

$$D=L\chi_{\parallel}^{-1}, \tag{53}$$

where L is the Onsager kinetic coefficient.[9] If L stayed finite at the stability limit, the diffusion constant would go to zero proportional to $\chi_{\parallel}^{-1}$, and the frequency given by Eq. (52) would indeed show the expected soft-mode behavior. For an isotropic Heisenberg ferromagnet in zero applied field above the Curie temperature, this argument has been shown to be invalid, and the diffusion constant is found to go to zero more slowly ($\propto\chi_{\parallel}^{-1/4}$), because L becomes singular at the transition.[9,10] For an anisotropic ferromagnet, however, L is expected to remain finite,[11] and the above argument should describe the softening of the spin-diffusion mode correctly.

For a more realistic model containing spin-phonon interactions, on the other hand, also the $q=0$ parallel component of the magnetization can relax, and one expects a soft parallel relaxation mode associated with the stability limit H_z^{c2}. In order to obtain a qualitative understanding of the dynamic behavior of such a system, we study here a simple model in which the effects of the spin-phonon interaction are approximated by Bloch-type relaxation terms[12] supplementing the RPA equation of motion of the Heisenberg Hamiltonian given in Eqs. (6)–(9). The equation of motion then takes the form of a modified Bloch equation:

$$\frac{d\boldsymbol{\sigma}}{dt}=\boldsymbol{\sigma}\times\mathbf{H}^{\mathrm{mol}}-\frac{1}{\tau_1}(\boldsymbol{\sigma}_{\parallel}-\boldsymbol{\sigma}_{\mathrm{eq}})-\frac{1}{\tau_2}\boldsymbol{\sigma}_{\perp}, \tag{54}$$

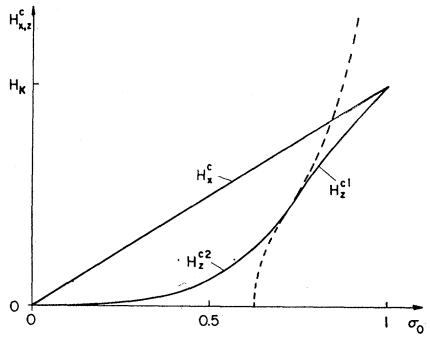

FIG. 5. Critical fields as function of the zero-field magnetization σ_0.

where we have assumed an isotropic g factor and put $\hbar=1$. The first term describes the precession of the magnetization around the instantaneous molecular field defined by Eq. (23), and the two last terms describe the relaxation of the magnetization $\boldsymbol{\sigma}$ towards the time-dependent equilibrium value $\boldsymbol{\sigma}_{\mathrm{eq}}=\sigma_{\mathrm{eq}}\mathbf{s}_{\mathrm{eq}}$ belonging to the instantaneous molecular field $\mathbf{H}^{\mathrm{mol}}$:

$$\boldsymbol{\sigma}_{\mathrm{eq}}=B_S(\beta SH^{\mathrm{mol}}),\quad \mathbf{s}_{\mathrm{eq}}=\mathbf{H}^{\mathrm{mol}}/H^{\mathrm{mol}}. \tag{55}$$

Different relaxation times τ_1 and τ_2 have been assumed for the components $\boldsymbol{\sigma}_{\parallel}$ and $\boldsymbol{\sigma}_{\perp}$ parallel and perpendicular to $\boldsymbol{\sigma}_{\mathrm{eq}}$. We introduce the projection operators

$$\mathbf{P}_{\parallel}=\mathbf{s}_{\mathrm{eq}}\mathbf{s}_{\mathrm{eq}},\quad \mathbf{P}_{\perp}=1-\mathbf{s}_{\mathrm{eq}}\mathbf{s}_{\mathrm{eq}} \tag{56}$$

such that

$$\boldsymbol{\sigma}_{\parallel}=\mathbf{P}_{\parallel}\cdot\boldsymbol{\sigma},\quad \sigma_{\perp}=\mathbf{P}_{\perp}\cdot\boldsymbol{\sigma}. \tag{57}$$

Equation (54) is the straightforward generalization of the form given by Wangsness[13] to the ferromagnetic case.

We calculate the dynamic susceptibility tensor $\chi(\omega)$ for this model in two steps: In the first step, we obtain the dynamic response of the magnetization to a perturbation of the molecular field, which is equal to the dynamic response of a free spin in an external field equal to $\mathbf{H}^{\mathrm{mol}}$. In the second step, we relate the change of the molecular field to the change of the external field.

We consider small dynamic deviations from equilibrium:

$$\begin{aligned}\mathbf{H}^{\mathrm{mol}}(t)&=\boldsymbol{\Lambda}_0+\delta\mathbf{H}^{\mathrm{mol}}e^{-i\omega t},\\ \boldsymbol{\sigma}(t)&=\boldsymbol{\sigma}_0+\delta\boldsymbol{\sigma}e^{-i\omega t},\\ \boldsymbol{\sigma}_{\mathrm{eq}}(t)&=\boldsymbol{\sigma}_0+\delta\boldsymbol{\sigma}_{\mathrm{eq}}e^{-i\omega t},\end{aligned} \tag{58}$$

where $\boldsymbol{\sigma}_0$ and $\boldsymbol{\Lambda}_0$ are the static equilibrium values of the magnetization and of the molecular field, respectively, which are related by Eq. (14). From now on, we drop the subscript 0 for the equilibrium properties. We obtain the linearized equation of motion

$$\begin{aligned}-i\omega\delta\boldsymbol{\sigma}=\delta\boldsymbol{\sigma}\times\boldsymbol{\Lambda}+\boldsymbol{\sigma}\times\delta\mathbf{H}^{\mathrm{mol}}\\ -(\tau_1^{-1}\mathbf{P}_{\parallel}+\tau_2^{-1}\mathbf{P}_{\perp})\cdot(\delta\boldsymbol{\sigma}-\delta\boldsymbol{\sigma}_{\mathrm{eq}}).\end{aligned} \tag{59}$$

According to Eq. (55), the change in $\boldsymbol{\sigma}_{\mathrm{eq}}$ is related to the

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change in H^{mol} by

$$\delta\boldsymbol{\sigma}_{\mathrm{eq}}=(1/S)\boldsymbol{\chi}_f\cdot\delta\mathbf{H}^{\mathrm{mol}}, \tag{60}$$

where $\boldsymbol{\chi}_f$ is the static free-spin susceptibility tensor given by Eqs. (28) and (29). We express the vector products in Eq. (59) in tensorial form with the help of the totally antisymmetric tensor $\boldsymbol{\varepsilon}$ of third rank, and solve for $\delta\boldsymbol{\sigma}$. The result can be written in the form

$$\delta\boldsymbol{\sigma}=(1/S)\boldsymbol{\chi}_f(\omega)\cdot\delta\mathbf{H}^{\mathrm{mol}}, \tag{61}$$

where $\boldsymbol{\chi}_f(\omega)$ is the dynamic free-spin susceptibility tensor

$$\boldsymbol{\chi}_f(\omega)=\chi_{f\parallel}(\omega)\mathbf{P}_\parallel+\boldsymbol{\chi}_{f\perp}(\omega)\cdot\mathbf{P}_\perp, \tag{62}$$

with

$$\begin{aligned}&\chi_{f\parallel}(\omega)=\chi_{f\parallel}(1-i\omega\tau_1)^{-1},\\ &\boldsymbol{\chi}_{f\perp}(\omega)=\chi_{f\perp}[(1-i\omega\tau_2)\mathbf{1}-\tau_2\Lambda\boldsymbol{\varepsilon}_\perp]^{-1}\cdot(\mathbf{1}-\tau_2\Lambda\boldsymbol{\varepsilon}_\perp).\end{aligned} \tag{63}$$

Here, $\boldsymbol{\varepsilon}_\perp$ is the totally antisymmetric tensor of second rank in the $\perp$ subspace defined by

$$\boldsymbol{\varepsilon}_\perp=\mathbf{s}\cdot\boldsymbol{\varepsilon}. \tag{64}$$

We now have to relate the change of the molecular field to the change $\delta\mathbf{H}$ of the external field. We write

$$\delta\mathbf{H}^{\mathrm{mol}}=S\boldsymbol{\lambda}\cdot\delta\boldsymbol{\sigma}+\delta\mathbf{H}, \tag{65}$$

where the first term represents the contribution induced by the change $\delta\boldsymbol{\sigma}$ of the magnetization. From Eq. (23) defining the molecular field, we find by using Eqs. (18), (25), and (29)

$$\boldsymbol{\lambda}=\boldsymbol{\chi}_f^{-1}-\boldsymbol{\chi}^{-1}. \tag{66}$$

By combining Eqs. (65) and (61), we obtain for the dynamic susceptibility tensor $\boldsymbol{\chi}(\omega)$ defined by

$$\delta\boldsymbol{\sigma}=(1/S)\boldsymbol{\chi}(\omega)\cdot\delta\mathbf{H} \tag{67}$$

the interesting relation

$$\boldsymbol{\chi}^{-1}(\omega)-\boldsymbol{\chi}^{-1}=\boldsymbol{\chi}_f^{-1}(\omega)-\boldsymbol{\chi}_f^{-1}. \tag{68}$$

With the help of Eqs. (62) and (63) it can be written in the form

$$\begin{aligned}\boldsymbol{\chi}^{-1}(\omega)=&\boldsymbol{\chi}^{-1}\\ &-i\omega[(\tau_1/\chi_{f\parallel})\mathbf{P}_\parallel+(\tau_2/\chi_{f\perp})(\mathbf{1}-\tau_2\Lambda\boldsymbol{\varepsilon}_\perp)^{-1}\cdot\mathbf{P}_\perp].\end{aligned} \tag{69}$$

The eigenfrequencies can be found as solutions of

$$\det\boldsymbol{\chi}^{-1}(\omega)=0. \tag{70}$$

The soft-mode behavior at a stability limit follows quite generally from the structure of Eq. (69): Whenever an eigenvalue of $\boldsymbol{\chi}^{-1}$ becomes zero, one of the eigenfrequencies goes to zero.

We apply these results to the case that the external field is in the easy direction, and the magnetization is antiparallel to the field. Then, the static susceptibility tensor has principal values $\chi_{xx}=\chi_{yy}=\chi_\perp$ and $\chi_{zz}=\chi_\parallel$ given by Eqs. (36) and (39). From Eq. (69), we find the principal values of the dynamic susceptibility tensor of the form

$$\chi_{+,-}(\omega)=\chi_\perp\Omega_{+,-}/(\Omega_{+,-}-\omega) \tag{71}$$

and

$$\chi_\parallel(\omega)=\chi_\parallel\Omega_\parallel/(\Omega_\parallel-\omega). \tag{72}$$

Equation (71) describes the dynamic response to perpendicular fields of left- and right-handed circular polarization (ferromagnetic resonance). The normal-mode frequencies are given by

$$\Omega_{+,-}=(\chi_{f\perp}/\chi_\perp)(\pm\Lambda-i\tau_2^{-1}). \tag{73}$$

According to Eq. (46) they go to zero as $(H_z^{c1}-H_z)$ at the stability limit H_z^{c1}. The dynamic response to parallel fields described by Eq. (72) has a purely imaginary eigenfrequency

$$\Omega_\parallel=-i(\chi_{f\parallel}/\chi_\parallel)\tau_1^{-1} \tag{74}$$

corresponding to a parallel relaxation mode. According to Eq. (47) it goes to zero as $(H_z^{c2}-H_z)^{1/2}$ at the stability limit H_z^{c2}. From the fluctuation-dissipation theorem it can be seen that the stability limits H_z^{c1} and H_z^{c2} are associated with critical fluctuation of the perpendicular and parallel components of the magnetization, respectively.

For fields applied in the hard direction which are larger than H_x^c, the static susceptibility has principal values $\chi_{xx}\neq\chi_{yy}\neq\chi_{zz}$. In this case, the perpendicular normal modes are elliptically polarized oscillations with eigenfrequencies

$$\begin{aligned}\Omega_{1,2}=\frac{\chi_{f\perp}}{(\chi_{yy}\chi_{zz})^{1/2}}\Bigg[&\pm\left(\Lambda^2-\tfrac{1}{4}\tau_2^{-2}\frac{(\chi_{yy}-\chi_{zz})^2}{\chi_{yy}\chi_{zz}}\right)^{1/2}\\ &-\frac{i}{2}i\tau_2^{-1}\frac{\chi_{yy}+\chi_{zz}}{(\chi_{yy}\chi_{zz})^{1/2}}\Bigg].\end{aligned} \tag{75}$$

As the field approaches the stability limit H_x^c, these frequencies become purely imaginary at a certain field $H_x>H_x^c$, and only one of them goes to zero at the stability limit. Thus, in this case, the soft mode is a perpendicular relaxation mode rather than a resonance mode. The parallel mode has an eigenfrequency

$$\Omega_\parallel=-i(\chi_{f\parallel}/\chi_{xx})\tau_1^{-1} \tag{76}$$

which stays finite for all $H_x>H_x^c$.

For fields applied at an arbitrary angle, the off-diagonal elements $\chi_{\parallel,\perp}$ of the static susceptibility couple the motions of the parallel and perpendicular components of the magnetization, and one obtains dynamic modes of a mixed character. In all cases, one mode becomes soft at the critical curve.

V. CONCLUDING REMARKS

We have obtained the stability limits of a uniaxial ferromagnet in the H_x-H_z plane for all temperatures up to the Curie temperature using the MFA, and we have discussed the critical dynamic behavior for a simple

model taking spin-lattice relaxation effects approximately into account. Of particular interest is the result that for fields applied in the easy direction in a temperature interval $T_1<T<T_C$, the instability occurs with respect to a change of magnetization magnitude. Associated with this transition are a soft parallel relaxation mode and critical fluctuations of magnetization magnitude.

For experimental observation of such behavior a very hard ferromagnet is required, because the temperature interval T_C-T_1 is of order $(K/J)T_C$. It has further to be noticed that the transition is of first order, and the ideal stability limit usually cannot be reached because of local nucleation of the reversed phase at imperfections, etc. It will, therefore, be necessary to infer the type of transition by an extrapolation procedure. If a suitable system can be found, it should be possible to study also the dynamic properties, because all relevant frequencies are in the microwave range or below.

The isotropic ferromagnet represents a singular case because $T_1\rightarrow T_C$ for $K\rightarrow 0$, and both the perpendicular and the parallel mode become soft at $T=T_C$. This may be the origin for some of the difficulties encountered in theoretical treatments of the phase transition of this system.

ACKNOWLEDGMENT

I am indebted to Dr. H. Rohrer for help with the numerical calculations.

PHYSICAL REVIEW VOLUME 187, NUMBER 2 10 NOVEMBER 1969

Direct Observation of Spin Rotation in $ErFeO_3$†

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(Received 6 May 1969)

We present conclusive evidence that the magnetic-spin reorientation occurring in $ErFeO_3$ at about 95°K proceeds by spin rotation.

IN a number of the weak ferromagnetic rare-earth orthoferrites (formula $RFeO_3$, where R is a rare earth), the direction of the magnetically ordered iron spins changes from the c to the a crystal axis on heating.[1–5] We report, in this article, a direct observation of the spin reorientation occurring in $ErFeO_3$ using the Mössbauer effect. In agreement with previous studies[4,5] our data provide conclusive evidence that the spins *rotate* continuously from the c to the a axis on heating.

Microscopic[2] and macroscopic[3] descriptions of the spin reorientation have indicated the possibility of two types of reorientation process:

(a) The spin magnetic moment *rotates* continuously from the c axis to the a axis, the rotation beginning at $T=T_1$, and ending at $T=T_2$, say.

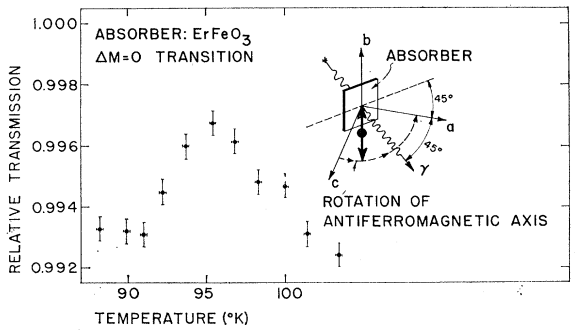

FIG. 1. Relative Mössbauer transmission of Fe^{57} in $ErFeO_3$ as a function of temperature. The source-absorber velocity was kept constant at the value corresponding to the peak of the $\Delta m=0$ absorption line. Below about 90°K, the spins lie along the c axis. On heating, the spins rotate towards the a axis. For $T\simeq 95$°K, the spins lie approximately parallel to the γ-ray direction, and the intensity of the $\Delta m=0$ line is reduced. A relative transmission of 1.000 corresponds to the transmission off resonance.

† Sponsored in part by the Air Force Materials Laboratory Research and Technology Division AFSC through the European Office of Aerospace Research, United States Air Force, under Contract No. F 61052-67C-0040.

* Part of a Ph.D. thesis to be submitted to the Feinberg Graduate School of the Weizmann Institute of Science.

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