Second-order phase transitions in ferromagnetic materials in weak fields near the Curie point

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A review is given of the present state of theoretical and experimental research on second-order phase transitions in anisotropic and nonuniformly magnetized ferromagnetic and ferrimagnetic materials and on phenomena observed near the Curie point in weak magnetic fields ($H < H_A, H_d$, where H_A and H_d are the anisotropy field and demagnetizing field, respectively). The nature of these transitions is examined, and it is shown that for anisotropic and nonuniformly magnetized ferromagnetic materials the Curie point is not an isolated point on the H-T plane. The experimental and theoretical data indicate the existence of a line of second-order phase transitions in a magnetic field applied in certain definite directions with respect to the anisotropy axis. This line of transitions is described by the law $T_c(H) = T_c(0)(1 - AH^{\omega})$; theoretical estimates in the molecular-field approximation yield values $\omega = 2$ for ferromagnetic materials of the easy-axis and easy-plane type and $\omega = 2/3$ for cubic ferromagnetic materials. The experimental results on the equilibrium properties (the magnetization, susceptibility, specific heat, magnetostriction, Faraday effect, etc.) and dynamic properties (the speed and attenuation rate of ultrasonic waves, the dynamic susceptibility) not only confirm the existence of a line of phase transitions but also indicate that spin fluctuations play a decisive role in the formation of the transition between ferromagnetic and paramagnetic phases in a weak magnetic field.

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CONTENTS

1. INTRODUCTION

The physics of phase transitions and critical phenomena has produced results of fundamental significance for elucidating the nature of the critical state of matter. Many of these results are based on studies of the critical anomalies for phase transitions of the ferromagnet-paramagnet type in the absence of an external magnetic field. Research on phase transitions in nonzero magnetic fields began comparatively recently.

In a magnetic field the magnetic structure of a paramagnetic material is not fundamentally different from that of a ferromagnetic one, for the symmetry in the spin distribution is the same in the two objects. The two phases become identical in their magnetic structure. Consequently, the magnetic field destroys the ferromagnet-paramagnet phase transition. It is for this reason that it is usually assumed that the Curie point or Curie temperature is an isolated point on the H-T diagram.[1,2] This assertion is valid only for ideal isotropic ferromagnetic materials of unbounded size. Real ferromagnetic materials, on account of both the anisotropic-exchange and relativistic interactions, are always anisotropic (have shape, magnetocrystallographic, and exchange anisotropy).

In nonideal ferromagnetic materials, as has been shown in studies over the past twenty years,[3-39] a phase transition occurs in the region of the Curie temperature even in the presence of a magnetic field smaller than the anisotropy and demagnetizing fields. Singular behavior of ferromagnetic materials in a nonzero magnetic field was first detected by Teaney, van der Hoeven and Moruzzi[3] in a study of the behavior of the specific heat of EuS in a magnetic field in the region of the Curie point. Those authors came to the conclusion that the critical point of the ferromagnetic europium sulfide is not an isolated point on the H-T diagram. It was subsequently pointed out by Arrott[4] that the behavior of the line on the H-T diagram of a ferromagnetic material under the experimental conditions of Ref. 3 can be explained by supposing that the uniformly magnetized ferromagnetic state is not the only possible ground state. Arrott reached this conclusion on the basis of the theory of Griffiths,[5] who considered a model ferromagnetic substance consisting of spins localized at lattice sites and interacting by exchange and dipole-dipole forces. A transition occurred from a ferromagnetic state with a nonuniform magnetization to a uniformly magnetized paramagnetic state. Arrott's idea was developed further by a number of authors.[6-13]

Phase transitions in anisotropic ferromagnetic materials in weak magnetic fields were studied in Refs. 7–24, in which the thermodynamic theory of Landau[14] was used to investigate the features of the magnetic

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properties not only of uniaxial magnetic materials but also of ferromagnetic materials having an easy axis along [100] or [111]. The main result of these studies—the occurrence of second-order phase transitions near the Curie point for certain definite directions of the magnetic field with respect to the axis of easy magnetization—was confirmed by theoretical calculations based on the scaling hypothesis[8,10,24,25] and on renormalization-group studies.[12,13,26,43-50]

The study of magnetic phase transitions at the Curie point in weak magnetic fields has thus led to a new field of scientific research which has already yielded fundamentally new and important results. These results include, first of all, the abrupt changes observed in the temperature dependence of the magnetic, magnetoelastic, magnetooptic, and other properties of ferromagnetic materials in weak magnetic fields. By studying these effects one can determine the spontaneous magnetization and spontaneous magnetostriction and obtain a number of critical exponents and amplitudes on the basis of kink phenomena, etc. These effects are of important interest not only for the further development of the theory of phase transitions but for practical applications as well.

In spite of the great interest currently shown in the study of phase transitions in ferromagnetic materials in weak magnetic fields, the experimental data are not as yet sufficient to confirm the theoretical deduction conclusively, and the existing experimental and theoretical results have not been generalized or compared. In the present review we have therefore undertaken to fill this gap and examine the features of the magnetic properties of isotropic and anisotropic ferromagnetic materials in weak magnetic field in the vicinity of the Curie point. Much of our attention is devoted to the effect of fluctuations of the magnetization on the formation of a second-order phase transition in a magnetic field. In addition, we consider the features of the nonmagnetic properties of ferromagnetic and ferrimagnetic materials, a subject that was first studied by the authors of this review and their co-workers.

2. PHASE TRANSITIONS IN ISOTROPIC FERROMAGNETIC MATERIALS IN AN EXTERNAL MAGNETIC FIELD

Griffiths[5] proved a theorem which states that a system of magnetic dipoles in a crystal lattice with magnetic dipole-dipole and exchange interactions has, in the thermodynamic (large-volume) limit, a definite bulk free energy for $H=0$ that does not depend on the shape of the sample. An immediate consequence of this theorem is that in the absence of an external magnetic field the lowest energy state of an isotropic magnetic system is not a state of uniform magnetization. To illustrate this conclusion Arrott[4] considered a model of a thin isotropic ferromagnetic toroid, for which the ground state in the absence of magnetic field is a nonuniformly magnetized mode. He showed that when the sample is magnetized by a weak magnetic field applied perpendicular to the plane of the toroid, a second-order phase transition occurs from the nonuniformly magnetized ferromagnetic state to a uniformly magnetized paramagnetic state, with the transition temperature depending quadratically on the strength of the magnetic field. Assuming that the lowest energy state of the system is a state with nonuniform magnetization, Arrott found the conditions under which the system undergoes a second-order phase transition in a magnetic field.[4]

Because of the dipole-dipole interaction, which gives rise to a demagnetizing field, the nonuniformly magnetized state of the toroid (the N state) is one in which the magnetization vector lies in the plane of the toroid. The uniformly magnetized state (the U state) is one in which the magnetization vector is parallel to a magnetic field applied perpendicular to the plane of the toroid. The problem of determining the conditions under which the N state converts to the U state is similar to the problem of determining the perpendicular susceptibility of an antiferromagnetic material.[2,6]

In a field $\mathbf{H}=H\mathbf{z}$ perpendicular to the plane of the toroid, the magnetization of a volume element will have the decomposition

$$\mathbf{M} = M\mathbf{m} = M_z\mathbf{z} + M_\theta\boldsymbol{\theta}, \tag{2.1}$$

where the unit vector $\mathbf{z}$ is perpendicular to the plane of the toroid and the unit vector $\mathbf{m}$ is tangential to the circumference in the plane of the toroid for the volume element under consideration. Then in the molecular-field approximation the total field acting on a localized spin is equal to the resultant of the external, exchange, and demagnetizing fields:

$$H_{\text{eff}} = H\mathbf{z} + \gamma M\mathbf{m} - DM_z\mathbf{z}. \tag{2.2}$$

The condition that there be no net torque acting on the magnetic moments ($\mathbf{M}\times\mathbf{H}_{\text{eff}}=0$) implies the equation

$$\mathbf{M}_\theta(H - DM_z) = 0. \tag{2.3}$$

This equation yields the conditions under which the U or N state is realized. For the U state $M_\theta=0$ and $M_z=M$. In this case measurements along H yield the usual temperature-dependent magnetization. For the N state one has $M_\theta\neq 0$ and $M_z=H/D$ and, consequently, the magnetization measured along the field H is proportional to the field and independent of the temperature, like the perpendicular susceptibility of an antiferromagnetic material.

The linear field dependence of the magnetization of finite ferromagnetic material samples was first noted by Néel,[15] and then Fallot[16] verified this dependence experimentally for many ferromagnetic materials. Subsequent experimental studies[17-23] of the magnetic properties of polycrystalline ferromagnetic and ferrimagnetic materials in weak magnetic fields ($H<H_d$) not only confirmed the linear dependence of M on H but also showed that the magnetization remained independent of temperature all the way up to $T=T_c(\mathbf{H})$ [M falls off sharply above this temperature]. This phenomenon, which in the ferrites has come to be called the "kink," was first observed by Kamilov[22] and used to determine T_c. Experimental studies[3] of the specific heat of EuS in the vicinity of T_c showed that the specific-heat peak

shifted to lower temperatures with increasing magnetic field.

These facts and considerations served as a basis for theoretical calculations carried out in the molecular-field approximation.

The stability regions of the U and N states can be found from the difference in the free energies of these states as functions of the temperature and external field. Wojtowicz and Rayl[6] gave the corresponding calculation for the case $S=1/2$. Here we shall present their derivation. In the U state the effective field is

$$H_{\text{eff}} = (H + \gamma M - DM)_z. \tag{2.4}$$

In the N state the external and demagnetizing fields are directed along the z axis, and therefore $H_{\text{eff}}=\gamma mM$. The corresponding free energies of the two states (in units of γM^2) are of the form

$$F_U = F_0(\tau) - \tau \ln\{2\operatorname{ch}[(h - d\sigma_U + \sigma_U)\tau^{-1}]\} - \frac{1}{2}d\sigma_U^2 + \frac{1}{2}\sigma_U^2, \tag{2.5}$$

$$F_N = F_0(\tau) - \tau \ln\left(2\operatorname{ch}\frac{\sigma_N}{\tau}\right) - \frac{1}{2}\frac{h^2}{d} + \frac{1}{2}\sigma_N^2, \tag{2.6}$$

where $h=H/\gamma M_0$, $d=D/\gamma$, M_0 is the magnetization at $T=0$, $\tau=T/T_c(0)$ is the reduced temperature, and $T_c(0)$ is the Curie temperature. The quantities $\sigma_N=M_N/M_0$ and $\sigma_U=M_U/M_0$ are taken as the order parameters. By minimizing F_U and F_N with respect to σ_U and σ_N at constant τ and H, we obtain

$$\sigma_U = \operatorname{th}[(h - d\sigma_U + \sigma_U)\tau^{-1}], \tag{2.7}$$

$$\sigma_N = \operatorname{th}\frac{\sigma_N}{\tau}. \tag{2.8}$$

The condition for a transition from state N to state U is the equality of the free energies ($F_N=F_U$), which corresponds to the condition

$$\sigma_U = \sigma_N = \frac{h}{d}. \tag{2.9}$$

The reduced temperature at which the transition from state N to state U occurs is denoted by $\tau_c(h)$, and the actual transition temperature is denoted by $T_c(H)$. Then it is easily shown from (2.7) and (2.9) that

$$\tau_c(h) = \frac{h}{d}\left(\operatorname{arcth}\frac{h}{d}\right)^{-1}. \tag{2.10}$$

In the case of weak magnetic fields expression (2.10) assumes the form

$$\tau_c(h) = 1 - \frac{1}{3}\left(\frac{h}{d}\right)^2. \tag{2.11}$$

For $T<T_c(H)$ we have $F_N<F_U$, and the nonuniformly magnetized state is stable; for $T>T_c(H)$ the parameter σ_N is smaller than h/d, and only the U state is stable. At the temperature $T_c(H)$ the magnetizations are equal: $M_N=M_U$. Above $T_c(H)$ the experimentally measured component of the magnetization is $M_z=M_0\sigma_U$, while below $T_c(H)$ it is $M_z=H/D$.

The temperature $T_c(H)$, which depends quadratically on the field H, thus separates states having nonuniform and uniform magnetization. This temperature has come to be called the "kink" temperature. Figure 1a shows the results of a numerical solution of equations (2.7) and (2.8) in various magnetic fields. As phase-transition points, the kink points $T_c(H)$ are not fundamentally different from $T_c(0)\equiv T_c$.

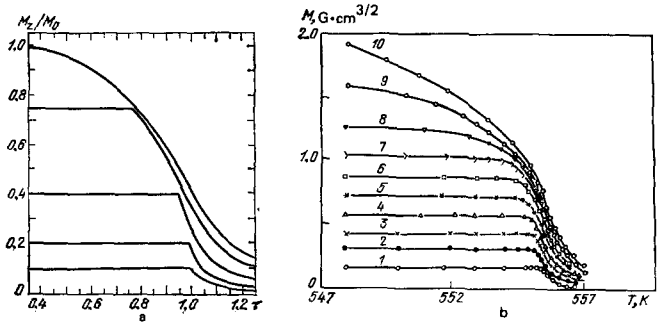

FIG. 1. Temperature dependence of the magnetization in weak magnetic fields. a) The theoretical curves[6] (in all cases $d=0.04$, for the upper curve $h/d=1$); b) the experimental data for the ferrite garnet $Dy_3Fe_5O_{12}$: 1) 4 Oe, 2) 6 Oe, 3) 8 Oe, 4) 10 Oe, 5) 12 Oe, 6) 13.5 Oe, 7) 16 Oe, 8) 20 Oe, 9) 30 Oe, 10) 40 Oe.

The case of arbitrary S was considered by Durczewski.[7] In the nonuniformly magnetized phase the magnetization components along the field (σ_N') and perpendicular to the field (σ_N'') obey the following laws:

$$\sigma_N' = \frac{h}{d}, \quad \sigma_N'' = \pm\sqrt{\sigma_N^2 - \left(\frac{h}{d}\right)^2}. \tag{2.12}$$

In the vicinity of the phase-transition temperature T_c one has for the case $S=1/2$

$$(\sigma_N'')^2 \approx 3\left(1 - \frac{T}{T_c(H)}\right) \tag{2.13}$$

and, consequently, the magnetization of the nonuniform mode goes continuously to zero at $T=T_c(H)$, confirming the presence of a second-order phase transition. The calculations of Durczewki showed that for temperatures near $T_c(H)$ one should observe anomalies not only in the specific heat but also in the weak-field susceptibility of the nonuniform mode, while the susceptibility in the field direction, according to (2.12), is constant:

$$\chi = \frac{1}{d}. \tag{2.14}$$

The laws (2.10)–(2.14) obtained in the molecular-field approximation[4,6,7] show that in isotropic ferromagnetic materials with dipole-dipole interactions the magnetic field does not destroy the second-order phase transition. The characteristic features of the transitions from the nonuniformly magnetized state to the uniformly magnetized state are thus: 1) the magnetization depends linearly on H; 2) the magnetization and susceptibility are independent of temperature in the nonuniformly magnetized phase all the way up to the temperature $T_c(H)$, which depends quadratically on H; 3) the magnetization and susceptibility of the nonuniform mode are anomalous at $T_c(H)$.

The large body of experimental data for polycrystalline and single-crystal samples qualitatively confirms these features of the magnetic properties.[17-22] As an example we show in Fig. 1b the data on the temperature dependence of the magnetization in various fields for the

ferromagnetic material $Dy_3Fe_5O_{12}$. However, the characteristic shortcoming of molecular-field theory—the neglect of fluctuations—leads to quantitative disagreement between theory and experiment. In particular, the line of second-order phase transitions in the H-T plane is described by a power-law dependence with critical exponent $\omega = 1/\beta$, and not by the quadratic dependence (2.11). Figure 2 shows our experimental data on the shift $T_c(H)$ in weak magnetic fields for the ferrite $Gd_3Fe_5O_{12}$; it is readily apparent that the deviation from a quadratic dependence grows as the temperature approaches $T_c(0)$. These data imply that fluctuations of the magnetization play an important role in the formation of the transition from the nonuniformly magnetized to the uniformly magnetized state in isotropic ferromagnetic and ferrimagnetic material with dipole-dipole forces. It must be noted that as yet there have been no theoretical papers taking fluctuations into account for nonuniformly magnetized ferromagnetic materials.

Nevertheless, the theoretically predicted and experimentally observed features of the magnetic properties in isotropic ferromagnetic materials of finite size are subjects of significant interest. In particular, it should be possible to use "kink" phenomena to determine accurately the temperature dependence of the spontaneous magnetization and also the Curie temperature of ferromagnetic and ferrimagnetic materials.

As is well known, many methods have been developed[1,2] for determining M_s and T_c. However, the problem of determining these parameters to high precision remains unsolved to this day. At the present time the techniques for determining M_s and T_c also include nuclear methods such as NMR, neutron diffraction, and the Mössbauer effect, which do not require the application of an external magnetic field. However, these methods also have certain shortcomings, chiefly that to determine M_s and T_c one must take into account the characteristic times governing the relaxation processes. As T_c is approached the relaxation time increases and can become equal to the characteristic experimental times for the nuclear techniques, this leads either to a smearing out of the phase-transition picture or to an overestimate of T_c, as, for example, in the case when superparamagnetism arises. In certain cases[28] nuclear methods also give an appreciably understated value of T_c. For this reason we believe that direct magnetic measurements yield more accurate values of M_s and T_c in spite of the fact that these measurements are made in finite magnetic fields.[22]

In making magnetic measurements one usually obtains from the experiment a set of M-H-T data points and extrapolates them to the field value $H=0$ to find M_s and T_c. At the present time several methods of extrapolation are used: the well-known Belov[29] method of thermodynamic coeffficients and an equivalent method die to Arrott,[30] and a modification of the Belov method developed by Popovici,[31] which heuristically makes allowance for fluctuations. These methods are described in sufficient detail in the literature, and we shall therefore discuss only the more recently developed kink method.

The kink method is based on a simple idea that stems from the considerations discussed above. When a ferromagnetic sample of finite size is located in a magnetic field H, a demagnetizing field $H_d = DM$ arises in the sample, and the internal field acting on the sample is given by the expression

$$H_i = H - DM. \tag{2.15}$$

In weak magnetic fields, as was shown above, the magnetization depends linearly on the field, so that the demagnetizing field is equal to the external field, and $H_i = 0$. It follows from (2.15) that in this case the maximum possible magnetization cannot be greater than H/D. On the other hand, the maximum magnetization corresponding to total magnetic saturation at temperature T is equal to the spontaneous magnetization $M_s(T)$. This circumstance makes it possible to determine $M_s(T)$. The breakdown of the condition $H_i = 0$ can be detected in two ways:

1) by varying the magnetic field at fixed temperature;

2) by varying the temperature at fixed field.

In the first case one experimentally constructs the magnetization isotherms. With increasing field H the magnetization of an isotropic ferromagnetic sample under the condition $\chi \gg 1/D$ will grow in proportion to the field H as long as $H < DM_s$. Above a certian critical field $H_k = DM_s$ the internal field H_i ceases to be zero and a kink appears on the isotherm. From the point of the kink one determines the value of M_s corresponding to the temperature of the isotherm in question (Fig. 3). To construct the temperature dependence of M_s one

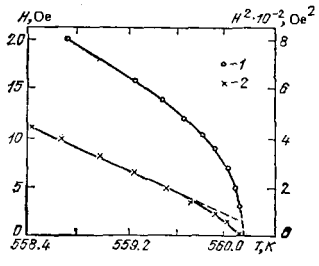

FIG. 2. Shift of Curie point[22] in weak magnetic fields for $Gd_3Fe_5O_{12}$. 1) The kink points on the H-T plane, 2) the points on the H^2-T plane.

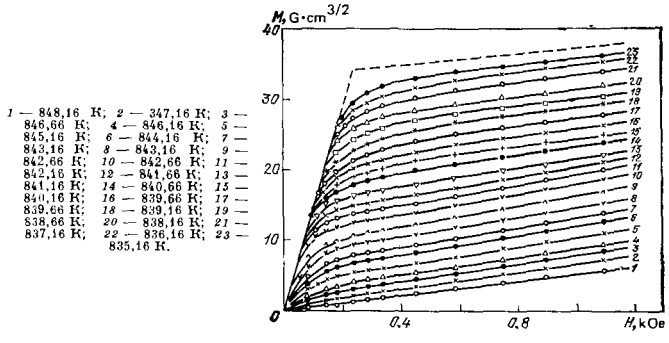

FIG. 3. Isotherms of the field dependence of the magnetization of Fe_3O_4 for various temperatures[22].

plots a set of isotherms. In real ferromagnetic materials the kink is not clearly expressed, as can be seen from our experimental magnetization isotherms for magnetite above T_c (Fig. 3). In the second case one records the temperature dependence of the magnetization for $H < H_d$. The field-induced magnetization will remain constant and equal to $M(T) = H/D$ as long as $M(T) < M_s(T)$. At a certain temperature $T = T_c(H)$ a sharp decrease in the magnetization (the kink effect) is observed. The condition $M(T) < M_s$ implies that the temperature region $T < T_c(H)$ corresponds to the phase with a uniform distribution of the magnetization. At the kink point one has $H_i = 0$, and $M = M_s$ when the temperature reaches the kink temperature. After the kink point the condition $H_i = 0$ is no longer satisfied. As the temperature is raised further the magnetization decreases, but at each temperature $M(T)$ it will be greater than M_s by an amount ΔM on account of the presence of a paraprocess in the external field, which is diminished by an amount $\Delta H_i = D\Delta M$. To construct $M_s(T)$ one plots a set of curves $M(T)$ for various fields H. Figure 1b shows the magnetization versus temperature curves for dysprosium ferrite garnet. The kink temperature is shifted to lower values with increasing H. At all temperatures $T > T_c(H)$ the field H produces a uniform magnetization of the ferromagnetic sample, and so this region corresponds to the uniformly magnetized phase. The kink point $T_c(H)$ is the point of the phase transition in the magnetic field.

Thus, by recording the curves $M(T)$ in various fields for samples with a regular geometric shape—spheres, thin disks, etc.—for which the demagnetizing factor D is known exactly, we have a set of points which determines the dependence of M_s on T. The temperature at which M_s goes to zero corresponds to the Curie temperature. For comparison we give in Fig. 4 the curves of $M_s(T)$ reconstructed by various extrapolation methods. Unlike the other methods, the kink method is not associated with any kind of assumptions (particular models, etc.) but is based on purely experimental facts, and therefore gives the most correct determination[22] of M_s and T_c.

It must be said that the theoretical formulation of the kink method based on molecular-field theory, despite the shortcomings of the latter, gives an adequate description of the general character of the second-order phase transition, characterized by the presence of a nonuniformly magnetized state, that is observed in isotropic ferromagnetic samples of finite size.

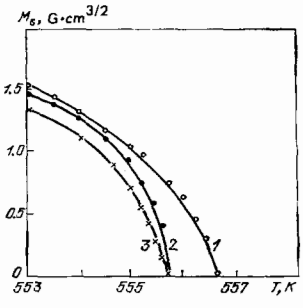

FIG. 4. Temperature dependence[22] of the spontaneous magnetization of $Dy_3Fe_5O_{12}$ reconstructed by various extrapolation methods. 1) kink method, 2) Belov method,[29] 3) Popovici method.[31]

In the papers of Durczewski[7] it was noted that the kink effect could be described without necessarily resorting to the hypothesis of a nonuniformly magnetized state in ferromagnetic materials, and that it was sufficient to consider anisotropy (in this case the shape anisotropy). However, neutron-diffraction experiments[32] have revealed nonuniformly magnetized regions near the Curie point in ferromagnetic samples of finite size, and so one should apparently differentiate between phase transitions in isotropic and anisotropic ferromagnetic samples in a magnetic field.

The nonuniformly magnetized state has so far been observed with the aid of neutrons in only a single study. At the same time, a ferromagnetic sample of finite size usually breaks up into domains. All our arguments concerning the transition from the nonuniform to the uniform state are also valid for the transition of a ferromagnetic sample to a uniform state from a state broken up into domains, which also occurs as a second-order phase transition. Thus curves similar to those given in Figs. 1 and 3 (and, incidentally, many subsequent curves as well) are obtained in all cases: 1) for the model of an anisotropic ferromagnetic material with a field perpendicular to the anisotropy axis, 2) for the transition from a nonuniform phase to a uniform phase, and 3) for the transition from a multidomain state to a uniform state. It has been pointed out by A. S. Borovik-Romanov that the last of these transitions should also be observed for a uniaxial ferromagnetic sample with a magnetic field directed along the easy axis.

3. PHASE TRANSITIONS IN ANISOTROPIC FERROMAGNETIC MATERIALS IN A MAGNETIC FIELD. MAGNETIC PROPERTIES

Following Refs. 8–11, we base our treatment of phase transitions in anisotropic ferromagnetic materials on the thermodynamic theory of Landau. In this approximation the free energy of an anisotropic ferromagnetic substance in the vicinity of T_c can be written in the form

$$F = F_0 + \sum_i a_i M_i^2 + \sum_{i,k} b_{ik} M_i^2 M_k^2 - \mathbf{H}\cdot\mathbf{M}, \tag{3.1}$$

where a_i and b_{ik} are thermodynamic coefficients, M_i are the components of the magnetization vector along the coordinate axes, and H is the magnetic field.

One can easily show that (3.1) describes all the possible cases of anisotropy:

$$1.\quad a_x = a_y = a,\quad a_x - a_z = K_1 > 0,\qquad b_{xx} = b_{yy} = b_{zz} = \tfrac{1}{2} b_{xy} = \tfrac{1}{2} b_{zx} = b \tag{3.2}$$

describes a ferromagnetic sample of the easy-axis type,

$$2.\quad a_x = a_y = a,\quad a_x - a_z = K_1 < 0,\qquad b_{xx} = b_{yy} = \tfrac{1}{2} b_{xy} = b,\quad b_{zz} = b_{zy} \tag{3.3}$$

describes a ferromagnetic sample of the easy-plane type.

$$3.\quad a_i=a,\quad b_{ii}=b,\quad b=\frac{1}{2}b_{ik}=K_2>0 \tag{3.4}$$

describes a cubic ferromagnetic sample with easy axis along [111], and

$$4.\quad a_i=a,\quad b_{ii}=b,\quad b=\frac{1}{2}b_{ik}=K_2<0\quad (i\neq k) \tag{3.5}$$

describes a ferromagnetic sample with easy axis along [100].

From the expression (3.1) for the free energy we find the conditions under which the second-order phase transition continues to exist in a magnetic field for the four cases of anisotropy.

For a uniaxial crystal one can, using (3.2), obtain the equilibrium values of the magnetization by minimizing (3.1) with respect to M_x and M_y. As a result, for the case when the anisotropy axis is along z and the field is directed along x, one finds

$$0=M_z[2(a-K_1)+4bM_x^2+4bM_z^2], \tag{3.6}$$

$$H_x=M_x[2a+4bM_x^2+4bM_z^2]. \tag{3.7}$$

Equation (3.6) has two solutions:

$$M_z=0, \tag{3.8}$$

$$M_z^2=-[(a-K_1)+2bM_x^2]\frac{1}{2b}. \tag{3.9}$$

The first solution corresponds to the paramagnetic phase, the second to the magnetic phase. The transition from one phase to the other occurs if

$$(a-K_1)+2bM_x^2=0. \tag{3.10}$$

From equation (3.7) it follows that M_x does not go to zero at any temperature. However, at the point corresponding to (3.10) there is a kink on the $M_x(T)$ curve. In fact, if $M_z=0$ and condition (3.10) is satisfied, it follows from (3.7) that

$$M_x=\frac{H_x}{2K_1}. \tag{3.11}$$

This solution corresponds to the minimum of the energy not only at the transition point but also throughout the entire ferromagnetic phase if

$$(a-K_1)+2bM_x^2<0, \tag{3.12}$$

while in the paramagnetic phase ($a-K_1+2bM_x^2>0$) M_x is determined by the equation

$$H_x=M_x(2a+4bM_x^2). \tag{3.13}$$

As is seen from Fig. 5, which shows the curves of the temperature dependence of M_x and M_z in different magnetic fields ($H<H_d$), M_z decreases continuously as $T\to T_c(H)$, while M_x remains constant up to the temperature at which the system undergoes a transition from the ferromagnetic to the paramagnetic state. In a transverse field, as follows from (3.8)–(3.11), a uniaxial ferromagnetic sample undergoes a second-order phase transition at a temperature

$$T_c(H)=T_c(0)-\frac{2b}{a^1}\left(\frac{H_x}{2K_1}\right)^2, \tag{3.14}$$

where $a^1>0$ is defined from the expansion $a-K_1=a^1(T-T_c)$. According to (3.14), $T_c(H)$ is shifted to lower temperatures with increasing H in accordance with a quadratic law, and the temperature $T_c(H)$ separates phases with $M_z\neq 0$ and $M_z=0$ (Fig. 6).

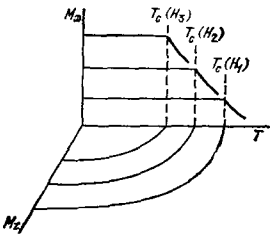

FIG. 5. Schematic curves for temperature dependence of M_x and M_z ($H_1<H_2<H_3$) for a uniaxial ferromagnetic material.[10]

Let us consider the behavior of the susceptibility in a uniaxial ferromagnetic sample. The susceptibility tensor has four components: χ_{zz}, $\chi_{xz}=\chi_{zx}$, and χ_{xx}. In a transverse field only the components χ_{xz} and χ_{xx} have meaning. For the ferromagnetic phase it can be shown from (3.8), (3.9), and (3.11) that

$$\chi_{xz}=\frac{H_x}{4K_1^2(a^1/2b)^{1/2}\sqrt{T_c(H)-T}}\sim[T_c(H)-T]^{-1/2}; \tag{3.15}$$

for $T\to T_c(H)$ we see that $\chi_{xz}\to\infty$, while

$$\chi_{xx}=\frac{1}{2K_1} \tag{3.16}$$

does not depend on T and H up to $T=T_c(H)$. In the paramagnetic phase we have $\chi_{xz}=0$ because $M_z=0$, while

$$\chi_{xx}=\frac{1}{2[a^1(T-T_c)+6bM_x^2+K_1]} \tag{3.17}$$

goes to infinity at the temperature

$$T=T_c-\frac{6bM_x^2}{a^1}-\frac{K_1}{a^1}. \tag{3.18}$$

It should be noted that the results obtained here are independent of the nature of the anisotropy and are valid for the case of both magnetocrystallographic and exchange anisotropy.

Despite the considerable number of theoretical studies of second-order phase transitions in uniaxial ferromagnetic materials,[8-11] experimental studies are still extremely rare. We know of only three studies[33-35] whose main purpose was to investigate the properties of uniaxial ferromagnetic materials in weak magnetic fields. Analysis of these papers shows that the easy-axis ferromagnetic substances $Cu(NH_4)_2Br_4\cdot 2H_2O$ (Refs.

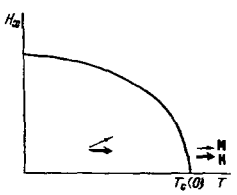

FIG. 6. Line of second-order phase transitions for a uniaxial ferromagnetic material.[10]

33 and 35), $CuK_2Cl_4 \cdot 2H_2O$ (Ref. 35), and MnP (Ref. 36) do in fact display a second order phase transition if the magnetic field is applied perpendicular to the easy axis. According to the experimental data obtained in the present study for the uniaxial ferromagnetic material Gd (shown in Fig. 7), in the ferromagnetic phase the magnetization increases linearly with H; the component M_x is independent of T in accordance with formula (3.11), while the component M_z goes to zero at $T = T_c(H)$. The transition is shifted to lower temperatures with increasing H in accordance with the law

$$T_c(H) = T_c - AH_x^{\omega}, \qquad (3.19)$$

where A is a constant independent of T and H, and ω has the values 2.5 ± 0.1 (Ref. 34) and 2.63 ± 0.1 (Ref. 33) for $Cu(NH_4)_2Br_4 \cdot 2H_2O$, 2.6 ± 0.1 (Ref. 35) for $CuK_2Cl_4 \cdot 2H_2O$, and 2.51 ± 0.1 (present study) for Gd.

According to the experimental data, the component of the susceptibility parallel to the field, depends weakly on T and H in the ferromagnetic phase, in agreement with (3.16), whereas in the paramagnetic phase near $T_c(0)$ it displays a maximum which shifts to higher temperatures with increasing H (Fig. 8).[34] For χ_{xz} there are as yet no direct experiments, but indirect data—in particular, the data on ultrasonic attenuation in MnP—show that χ_{xz} is singular at $T = T_c(H)$.

All these data indicate that the second-order phase transition in uniaxial ferromagnetic samples in a transverse field can be described qualitatively on the basis of Landau theory, but the lack of quantitative agreement between theory and experiment indicates that it is necessary to make allowance for the fluctuations of the easy-axis component of the magnetization.

Fluctuations were first incorporated into the framework of the Landau theory for uniaxial ferromagnets by Sznajd,[10] who showed that the correlation length of fluctuations of the easy-axis component of the magnetization in a transverse field has the same divergence at $T_c(H)$ as is found in the Ornstein-Zernike theory (see Ref. 14).

In ferromagnetic materials of the easy-plane type the free-energy expansion (3.1) will include all three components of the magnetization, and $K_1 < 0$. If the magnetic field is applied perpendicular to the easy

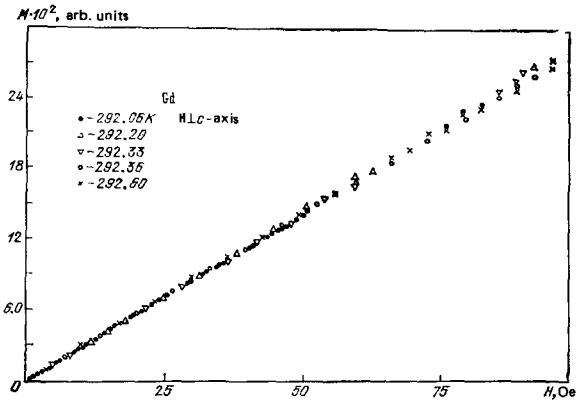

FIG. 7. Dependence of M_x on H for Gd at various T.

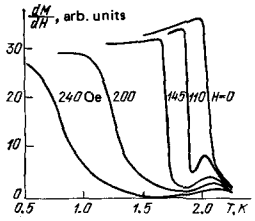

FIG. 8. Temperature dependence of the susceptibility[34] for $Cu(NH_4)_2Br_4 \cdot 2H_2O$ in a magnetic field at $p = 7.5$ kbar.

plane, M_x and M_y will display critical behavior, while M_z will not go to zero at any finite temperature. Consequently, the temperature and field dependence of the magnetization and susceptibility are the same as in uniaxial ferromagnets. However, as the calculations of Sznajd[13] and of Nikitin *et al.*[37,38] show, the line of second-order phase transitions in the H-T plane is given by the expression

$$H = \sqrt{A - T_c(H)}\,[T_c(0) - T_c(H)], \qquad (3.20)$$

which implies that there is a region of magnetic fields in which $T_c(H)$ grows with increasing H (see Fig. 9). In sufficiently weak fields the line of phase transitions is described by the same law as in the case of uniaxial ferromagnets. Experimental studies[37,38] of the magnetic properties of Tb_xGd_{1-x} alloys have shown that indeed $T_c(H)$ is shifted to lower temperatures with increasing H for fields up to 12 kOe.

As in ferromagnetic materials of the easy-axis and easy-plane type, second-order phase transitions are possible in cubic multiaxial crystals for a suitable choice of direction of the magnetic field with respect to the crystallographic axes. Theoretical studies based on Landau theory have been carried out for ferromagnetic materials with axes of easy magnetization along [100] and [111] in fields H ∥ [111], [110], and [100] (Refs. 12 and 40) and in fields H with arbitrary direction in the (100) plane.[10-13] These studies showed that second-order phase transitions occur in weak magnetic fields only in cases where the magnetic field lies at an angle $\pi/4$ to the two easy axes in the plane formed by these axes or is parallel to the hard axis (crystals with easy axis [100]). In a ferromagnetic material with easy axis along [111] the second-order phase transition is not destroyed by a weak magnetic field lying in the plane formed by the two hard axes or for H ∥ [100].

According to the thermodynamic theory of Landau, the free energy of a cubic ferromagnetic material near the Curie point in the limit of weak magnetic fields can be written in the form

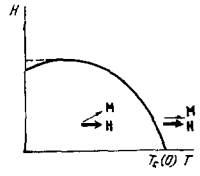

FIG. 9. Phase diagram of a ferromagnetic material of the easy-plane type.

$$F = F_0 + a^1 (T - T_c) M_1^2 + bM_1^2 M_k^2 + K_2 (M_x^4 + M_y^4 + M_z^4) - \mathbf{H}\cdot\mathbf{M}. \quad (3.21)$$

Analysis of this expression for the necessary conditions for the existence of a minimum implies that

$$2M_i [a^1 (T - T_c) + 2bM^2 + 2K_2 M_i^2] = H_i, \quad (3.22)$$

while sufficient conditions for a minimum are given by the inequalities

$$|\Delta| = \begin{vmatrix} f_{xx} & f_{xy} & f_{xz} \\ f_{yx} & f_{yy} & f_{yz} \\ f_{zx} & f_{zy} & f_{zz} \end{vmatrix} > 0, \quad \begin{vmatrix} f_{xx} & f_{xy} \\ f_{yx} & f_{yy} \end{vmatrix} > 0, \quad f_{xx} > 0, \quad (3.23)$$

where

$$f_{ii} = \frac{\partial^2 F}{\partial M_i^2} = 2 [a^1 (T - T_c) + 2bM^2 + 2 (2b + 3K_2) M_i^2], \quad (3.24)$$

$$f_{ik} = \frac{\partial^2 F}{\partial M_i \, \partial M_k} = 8bM_i M_k. \quad (3.25)$$

Let us consider first the case of a ferromagnetic sample with easy axis along [111], which is described by (3.21) with $K_2 > 0$. We apply an external magnetic field in such a way that its direction lies in the xy plane. Under these conditions equation (3.22) implies that

$$M_z^2 = -\frac{a^1 (T - T_c) + 2b (M_x^2 + M_y^2)}{2 (b + K_2)}, \quad (3.26)$$

while M_i is determined by the equation

$$4M_i (bM_i^2 + K_2 M_i^2) = H_i, \quad i = x, y. \quad (3.27)$$

These solutions satisfy the conditions for a minimum if

$$a^1 (T - T_c) + 2b (M_x^2 + M_y^2) < 0.$$

In the opposite case the conditions for a minimum are satisfied by the solutions

$$M_z = 0, \quad (3.28)$$

$$2M_1 [a^1 (T - T_c) + 2bM^2 + 2K_2 M_1^2] = H_1. \quad (3.29)$$

The states described by expressions (3.26), (3.27) and (3.28), (3.29) for a ferromagnetic material with easy axis along [111] will have the same energy under the condition

$$a^1 (T - T_c) + 2b (M_x^2 + M_y^2) = 0. \quad (3.30)$$

Substituting (3.28) and (3.29) into this equation, we obtain an expression for the temperature at which the phase transition occurs:

$$T_c (H) = T_c - \frac{2b}{a^1} \left(\frac{1}{2K_2}\right)^{2/3} (H_x^{2/3} + H_y^{2/3}). \quad (3.31)$$

It follows from expressions (3.26), (3.30), and (3.31) that M_z goes continuously to zero as $T \to T_c(H)$, indicating the presence of a second-order phase transition. To determine conclusively the order of the phase transition, let us consider the behavior of the susceptibility.

The susceptibility tensor of a ferromagnetic material with easy axis along [111] is given by the relation

$$\chi_{ik} = \frac{A_{ik}}{|\Delta|}, \quad (3.32)$$

where A_{ik} is the corresponding minor of the matrix $|\Delta|$ defined in (3.23). Analysis of the six components of the susceptibility shows that as $T \to T_c^*(H)$ (from the high-temperature side) only the component χ_{zz} is singular, going to infinity according to the power law

$$\chi_{zz} \sim [T - T_c (H)]^{-1}. \quad (3.33)$$

The remaining components χ_{ik} have finite values at $T = T_c$, except for $\chi_{xz} = \chi_{yz}$, which are equal to zero.

Thus, the transition from the state (3.26), (3.27) to the state (3.28), (3.29) is a second-order phase transition, since M_z changes continuously and χ_{zz} diverges. The lines of second-order phase transitions are described by equation (3.31) and can be represented on the $H-T$ diagram as the curves shown in Fig. 10.

In a cubic ferromagnetic material with axis of easy magnetization along the [100] direction ($K_2 \langle 0, b \rangle |K_2|$), second-order phase transitions are predicted for two directions of the magnetic field:

1. H lies in the xy plane at an angle of $\pi/4$ to the x and y axes.

2. $\mathbf{H} \parallel [111]$ (the hard axis).

Let us consider the first case. For convenience we introduce a new coordinate system obtained from the old by the transformations

$$X \to \frac{\sqrt{2}}{2} x + \frac{\sqrt{2}}{2} y, \quad Y \to -\frac{\sqrt{2}}{2} x + \frac{\sqrt{2}}{2} y, \quad Z \to z. \quad (3.34)$$

In the new coordinate system $H_y = H_z = 0$, while $H_x \neq 0$, and from (3.21) we obtain three types of solutions:

1. $M_y = M_z = 0, \quad 2M_x [a^1 (T - T_c) + (2b + K_2) M_x^2] = H_x;$ (3.35)

2. $M_y = 0, \quad M_z^2 = -\frac{[a^1 (T - T_c) + 2bM_x^2]}{2 (b + K_2)},$

$$\frac{2K_2 M_x}{b + K_2} [a^1 (T - T_c) + 3 (b + K_2) M_x^2] = H_x, \quad (3.36)$$

3. $M_z = 0, \quad M_y^2 = -\frac{[a^1 (T - T_c) + (2b + 3K_2) M_x^2]}{2b + K_2},$

$$\frac{4 |K_2| M_x}{2b + K_2} [a^1 (T - T_c) + 4 (b + K_2) M_x^2] = H_x. \quad (3.37)$$

As we see from (3.35), the stable state corresponds to a phase with $M_x^{(1)} \parallel H_x$ when $a^1 (T - T_c) + (2b + 3K_2) M_x^2 > 0$. In the phase corresponding to solutions (3.36) and (3.37), the magnetization does not coincide with the field direction. The conditions for a minimum imply that for

$$a^1 (T - T_c) + 3 (2b + K_2) M_x^2 < 0$$

the phases described by solution (3.36) or (3.37) can be stable, and for

$$a^1 (T - T_c) + 3 (2b + K_2) M_x^2 = 0 \quad (3.38)$$

a phase transition of first order arises between these states, since it is accompanied by a jump in the magnetization (Fig. 11). If, on the other hand,

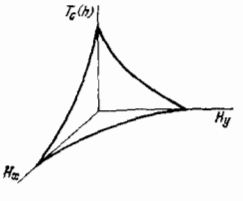

FIG. 10. Line of second-order phase transitions of cubic four-axis ferromagnetic materials.

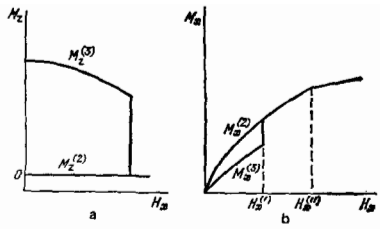

FIG. 11. Field dependence of M for $H \| x$ axis. a) M_z component of the magnetization, parallel to the easy axis; b) M_x component, parallel to H.

$a^1(T-T_c)+3(2b+K_2)M_x^2>0$ and $a^1(T-T_c)+(2b+3K_2)M_x^2<0$,

then only the phase described by equation (3.37) is stable. In this case a phase transition can occur to the state (3.35) when

$$a^1(T-T_c)+(2b+3K_2)M_x^2=0, \tag{3.39}$$

corresponding to a temperature

$$T_c(H)=T_c-\frac{3b+3K_2}{a^1}\left(\frac{H_x}{4|K_2|}\right)^{2/3}. \tag{3.40}$$

At this temperature there is no jump in the magnetization, as can be seen from Fig. 11b, and it follows from a calculation for the analogous case of a ferromagnetic material with easy axis along [111] that the susceptibility component χ_{yy} diverges as $T \to T_c(H)$:

$$\chi_{yy}=\{2[a^1(T-T_c)+(2b+3K_2)]M_x^2\}^{-1} \to [T-T_c(H)]^{-1}. \tag{3.41}$$

Consequently, for $T=T_c(H)$ one observes a phase transition of the second order. The phase diagram for this case is shown in Fig. 12.

In the case when $\mathbf{H} \| [111]$ the corresponding analysis shows that only a second-order phase transition is possible, at the temperature

$$T_c(H)=T_c(0)-\frac{b+|K_2|}{2a^1}\left(\frac{3H}{|K_2|}\right)^{2/3}. \tag{3.42}$$

Thus in cubic multiaxial ferromagnetic materials in weak magnetic fields a second-order phase transition is observed for a certain definite orientation of H with respect to the crystallographic directions. In contrast to the case of uniaxial ferromagnetic materials, for which the magnetization depends linearly on the field and remains independent of T, for multiaxial crystals

$$M_i=\frac{1}{4|K_2|}(H_i)^{1/3}, \quad (i=x,\ y). \tag{3.43}$$

Moreover, M_x and M_y are temperature dependent,

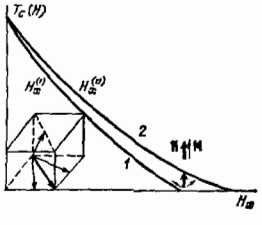

FIG. 12. Phase diagram of a three-axis ferromagnetic material. 1) Line of first-order phase transitions; 2) line of second-order phase transitions.

since for $H=0$ there exists a nonzero component M_i. As the temperature is increased this component decreases in absolute value, and at the transition point a kink appears on the $M_i(T)$ curve.

The general laws obtained from Landau theory for the temperature and field dependence of the magnetization and susceptibility in the case of multiaxial ferromagnetic materials have not as yet been confirmed experimentally, but it is clear that here too it will be necessary to take the critical fluctuations of the magnetization into account when considering second-order phase transitions in weak magnetic fields.

4. SCALING DESCRIPTION OF THE CRITICAL BEHAVIOR OF ANISOTROPIC FERROMAGNETIC MATERIALS

In the preceding paragraphs it was shown in the mean-field approximation that the second-order phase transition at T_c in anisotropic ferromagnetic materials is not destroyed by a weak magnetic field applied in a certain definite direction with respect to the anisotropy axis. The magnetization in this direction fluctuates strongly. The fluctuations can be taken into account phenomenologically in the approximation of scaling theory,[41] which postulates that the equilibrium properties in the region of a second-order phase transition temperature can be described by simple power laws. These power laws are given in Table I.

In Table I the quantities G and $\overline{G}$ are thermodynamic potentials whose specific form will be given later, and

$$\varepsilon=\frac{T-T_c(h)}{T_c(0)}=\varepsilon_0+\varepsilon_h, \quad \varepsilon_0=\frac{T-T_c(0)}{T_c(0)}. \tag{4.1}$$

Many of the power laws given in this table have been confirmed experimentally for uniaxial[17-22] and easy-plane[37,38] type ferromagnetic materials. However, the theoretical values of the critical exponents obtained by mean field theory differ appreciably from the experimental data (Table II).

The phase transitions in anisotropic ferromagnetic materials were first examined on the basis of scaling theory by Riedel and Wegner.[25] Concrete relations between the critical exponents were obtaind by Klamut and Sznajd[8,22] and Gaunt and Baker[40] using the Kadanoff

TABLE I.

Equilibrium properties	$h_\perp=h=0$, $\varepsilon \to 0$	$\varepsilon=0$, $h_\perp=0$, $h \to 0$	$\varepsilon=0$, $h=0$, $h_\perp \to 0$
1. $M=-(\partial G/\partial h)$	$-\varepsilon^{\beta}$	$\sim h^{1/\delta}$	
2. $\chi=(\partial M/\partial h)$	$\sim \varepsilon^{-\gamma}$	$\sim h^{-\lambda}$	
3. $s_h=-(\partial G/\partial \varepsilon)_h$	$\sim \varepsilon^{\psi}$	$\sim h^{-\varphi}$	
4. $c_h=T(\partial S_h/\partial \varepsilon)_h$	$\sim \varepsilon^{-\alpha}$	$\sim h^{-\alpha_h}$	
5. $M_\perp=(\partial \overline{G}/\partial h_\perp)$	$=0$		$\sim h_\perp^{1/\delta_\perp}$
6. $\chi_\perp=(\partial M/\partial h_\perp)$	$\sim \varepsilon^{-\gamma_\perp}$		$\sim h_\perp^{-\lambda_\perp}$
7. $s_{h\perp}=-(\partial \overline{G}/\partial \varepsilon)_{h_\perp}$	$\sim \varepsilon^{\psi}$		$\sim h^{\varphi_\perp}$
8. $c_{h\perp}=T(\partial S/\partial \varepsilon)_{h_\perp}$	$\sim \varepsilon^{-\alpha}$		$\sim h^{\alpha_\perp}$
9. $\varepsilon_h=\frac{T_c(0)-T_c(h)}{T_c(0)}$			$\sim h_\perp^{\omega}$

TABLE II.

Ferromagnetic material	Mean field theory			Experiment	
	Isotopic	Uniaxial	Multiaxial	Isotopic	Uniaxial
Critical exponents					
α	0	0	0		
β	1/2	1/2	1/2		
γ	1	1	1		
δ	3	3	3	4.6[22]	6.75[30]
$\delta_\perp$	1	1	3	1[10]	1[30]
ω	2	2	2/3	2.63[22]	2.53—2.63
$\gamma_\perp$	1				
ψ					
λ					
$\lambda_\perp$	0	0	0		
α_h	0	0	0		
$\alpha_\perp$					
φ	2/3	2/3	2/3		

representation.[41] On the basis of the ideas developed in these papers, let us consider how scaling theory is applied to the description of anisotropic ferromagnetic materials in weak magnetic fields.

First of all, let us discuss the relations between the scaling parameters and the critical exponent ω. In particular, from the scaling transformations of Kadanoff[41]

$$\tilde{\varepsilon} \sim L^y \varepsilon, \quad \tilde{h} \sim L^x h \tag{4.2}$$

it is easy to show that

$$\omega = \frac{x}{y}. \tag{4.3}$$

The basic assumption of scaling theory implies that

$$F(L^x h_\perp, L^y \varepsilon) = L^d (\varepsilon, h_\perp), \tag{4.4}$$

which is valid for any L. After transforming expression (4.4) to the form

$$F(\varepsilon, h_\perp) = \varepsilon^{\alpha/y} f(\varepsilon, h_\perp^{-y/x}), \tag{4.5}$$

differentiating (4.5) with respect to $h_\perp$, and taking into account that M is finite and $M_\perp \neq 0$ for $h_\perp = 0$, we find that the field dependence of $M_\perp$ for $\varepsilon = 0$ is given by the relation

$$M_\perp \sim h^{(d-x)/x} \equiv h_\perp^{1/\delta_\perp}. \tag{4.6}$$

This relation implies the inequality

$$\delta_\perp = \frac{x}{d-x}. \tag{4.7}$$

If we now use the familiar equality[41]

$$\beta = \frac{d-x}{y}, \tag{4.8}$$

and eliminate x and y from (4.3), (4.7), and (4.8), we obtain the new scaling relation

$$\omega \delta_\perp \beta = 1, \tag{4.9}$$

which was first derived by Klamut and Sznajd.[8]

Differentiating (4.4) with respect to $h_\perp$, one can obtain an equation relating ω with the critical exponent for the field dependence of the entropy at $\varepsilon = 0$:

$$\delta_\perp (\omega - 1 + \varphi_\perp) = 1. \tag{4.10}$$

Other relations between the critical exponents characterizing the critical behavior of an anisotropic ferromagnetic material have been obtained by Sznajd[24] using the parametric representation of Schofield.[42] This was done by introducing the new variables r, ϑ, and θ, which are related to the variables h, $h_\perp$, and ε (the field parallel and perpendicular to the axis and the temperature, respectively) by

$$\begin{aligned} h &= r^{h_1} \vartheta^{2h_2} \theta, \\ h_\perp &= r^{h'_1} \vartheta^{2h'_2} (1 - \theta), \\ \varepsilon &= r(1 - a\vartheta^2) + r\vartheta^2 \delta_{\theta,0}. \end{aligned} \tag{4.11}$$

We choose the thermodynamic potential in the form

$$G(h, \varepsilon) = F(h, \varepsilon) - F(0, 0) - Mh. \tag{4.12}$$

Let us consider two cases: 1) magnetic field directed along the anisotropy axis, i.e., $\theta = 1$, and 2) magnetic field perpendicular to the easy axis ($\theta = 0$). In the new variables the thermodynamic potentials for these two cases are, respectively:

$$G(r, \vartheta) = r^l g(\vartheta), \tag{4.13}$$

$$\bar{G}(r, \vartheta) = r^{l'} \bar{g}(\vartheta). \tag{4.14}$$

Then, using a familiar technique,[41] one can obtain the following relations among the critical exponents (scaling laws):

1. $\alpha + 2\beta + \gamma = 2$,
2. $\beta(\delta - 1) = \gamma$,
3. $\psi = 1 - \alpha$,
4. $\psi = \beta\varphi\delta$,
5. $\gamma = \beta\delta\lambda$,
6. $\alpha = \beta\delta\alpha_h$,
7. $\beta\delta_\perp\omega = 1$,
8. $\lambda_\perp + \beta\omega = 1$,
9. $\varphi_\perp + \omega(1 - \beta) = 1$,
10. $\varphi_\perp - \alpha_\perp = \omega$,
11. $-\omega(\gamma_\perp - 2 + \alpha) = 2$,
12. $\delta_\perp(1 - \lambda_\perp) = 1$,
13. $\delta_\perp(2\varphi_\perp - \alpha_\perp - 1) = 1$.

Relations 1—6 were obtained by Gaunt and Baker[40] and hold for isotropic ferromagnetic materials. The remaining relations 7–13 among the critical exponents characterize the critical behavior of anisotropic or nonuniformly magnetized ferromagnetic materials. Of these relations, 7, 8, and 12 hold in mean field theory (see Table II). The validity of the other relations cannot be ascertained owing to the lack of theoretical values of the critical exponents $\varphi_\perp$ and $\alpha_\perp$. Unfortunately, there have been no experimental checks of the scaling relations for anisotropic ferromagnetic materials, since of all the critical exponents so far only ω and $\delta_\perp$ have been determined (see Table II).

5. PHASE TRANSITIONS IN ANISOTROPIC FERROMAGNETIC MATERIALS IN THE RENORMALIZATION-GROUP APPROXIMATION

The renormalization-group method, which provides a microscopic justification for scaling theory and is based on the Kadanoff transformations,[41] has been applied to the description of phase transitions in uniaxial[12-43] and tetragonal[44] crystals in a magnetic field applied perpendicular to the easy axis. In cubic ferromagnetic materials this theory has been applied to the study of phase transitions in crystals with easy axis along [100] (with H lying in the plane formed by the two hard axes and along [111])[44,45] and with easy axis along

[111] (for $\mathbf{H} \| [100]$).[12,44] It was shown in these papers that in anisotropic ferromagnetic materials there is a second-order phase transition for the corresponding choices of the magnetic-field direction.

The general theory of second-order phase transitions in anisotropic ferromagnetic materials for $\mathbf{H} \neq 0$ was analyzed in the renormalization-group approximation by Ritter and Sznajd.[26] On the basis of the three-component vector model those authors showed that an anisotropic ferromagnetic sample in an external field can have either one or two critical variables, since the variable coinciding with the field direction does not go to zero at any finite temperature. The problem thereby reduces to one of considering the behavior of the Ising model (for the case of one variable) or the XY model (for the case of two variables) in a transverse magnetic field.

The existence condition for a second-order phase transition is found from the Landau-Ginzburg-Wilson Hamiltonian,[46] which has the following form in an external field:

$$\mathcal{H} = -\frac{1}{2}\sum_{i=1}^{3}\int_{q}(r_i^0+q^2)\,\sigma_q^i\sigma_{-q}^i + \sum_{i=1}^{3}h^i\sigma_0^i - \sum_{i,\,j=1}^{3}u_{ij}^0\int_{q}\int_{q_1}\int_{q_2}\sigma_q^i\sigma_{q_1}^i\sigma_{q_2}^j\sigma_{-q-q_1-q_2}; \tag{5.1}$$

here σ_q^i is a three-component classical spin with wave vector q, and

$$r_i^0 = a_i(T - T_c^i), \tag{5.2}$$

where T_c^i is the critical temperature for σ^i at $H=0$. By suitably choosing the interaction parameters r_i^0 and u_{ij}^0 in the Hamiltonian (5.1) we clearly can describe all types of anisotropy in ferromagnetic materials:

1. $r_1^0 = r^0$; $u_{ij}^0 = u^0 > 0$ — isotropic ferromagnetic material,
2. $r_1^0 \neq r_2^0 = r_3^0$; $u_{ij}^0 = u^0 > 0$ — uniaxial ferromagnetic material,
3. $r_1^0 \neq r_2^0 \neq r_3^0$; $u_{ij}^0 = u^0 > 0$ — orthorhombic ferromagnetic material,
4. $r_1^0 = r^0$, $u_{ij}^0 = u^0 > 0$, $i \neq j$; $u_{ij}^0 = u^0 + v^0$, $i = j$ — cubic ferromagnetic material,
5. $r_1^0 \neq r_2^0 = r_3^0$, $u_{ij}^0 = u^0 > 0$, $i \neq j$; $u_{ij}^0 = u^0 + v^0 > 0$, $i = j$ — tetragonal ferromagnetic material. (5.3)

Using the transformation

$$\sigma_q^i \to \sigma_q^i + M_i\delta_q, \tag{5.4}$$

and then a rotation of the spin component s_{q1} about the direction specified by the polar angles, we obtain from (5.1) the Hamiltonian in the new spin variables:

$$\mathcal{H} = -\frac{1}{2}\int\sum_{i,\,j}(r_{ij}+q^2\delta_{ij})\,s_q^i s_{-q}^i - \int_{q}\int_{q_1}\sum_{i,\,j}w_{ij}s_q^i s_{q_1}^i s_{-q-q_1} - 2w\int_{q}\int_{q_1}S_q^{(1)}S_{q_1}^{(2)}S_{-q-q_1}^{(3)} - \int_{q}\int_{q_1}\int_{q_2}\sum_{i,\,j}u_{ij}s_q^i s_{q_1}^i s_{q_2}^j s_{-q-q_1-q_2}^j - 4\int_{q}\int_{q_1}\int_{q_2}\sum_{i\neq j}v_{ij}s_{q_2}^i s_{q_1}^i s_{q_2}^j s_{-q-q_1-q_2}^j - \int_{q}\int_{q_1}\int_{q_2}\sum_{i\neq j\neq k}m_{ijk}s_q^i s_{q_1}^i s_{q_2}^j s_{-q-q_1-q_2}^j, \tag{5.5}$$

where w_{ij} and m_{ijk} are the new interaction constants.

Let us examine the conditions for the existence of a second-order phase transition in the XY model. We shall assume that the component $s_q^{(1)}$ does not display critical behavior, i.e., this component is in the direction of the magnetic field. Then $s_{q2}^{(2)}$ and $s_q^{(3)}$ will be the critical variables. Using the Wilson–Fisher-Nelson formalism,[47] one can reduce Hamiltonian (5.5) to the well-known form

$$\mathcal{H}_{XY} = -\frac{1}{2}\int_{q}\sum_{i=2}^{3}(r_{ij}+q^2)\,s_q^i s_{-q}^i - \int_{q}\int_{q_1}\int_{q_2}\sum_{i,\,j=2}^{3}\bar{u}_{ij}s_q^i s_{q_1}^i s_{q_2}^j s_{-q-q_1-q_2}^j, \tag{5.6}$$

where

$$\bar{u}_{ii} = u_{ii} - \frac{w_{1i}^2}{2r_{11}}, \quad \bar{u}_{23} = \bar{u}_{32} - \frac{2w^2 + w_{12}+w_{13}}{2r_{11}}. \tag{5.7}$$

The critical properties of the magnetic material described by the Hamiltonian (56) have been studied by many authors,[43-50] and it has been shown that the fixed point of the XY type is stable if

$$r_{11} > r_{22} = r_{33} \tag{5.8}$$

for any values of h. In addition, as was shown by Wilson and Fisher,[46] stability of this fixed point requires satisfaction of the conditions:

$$r_{ij} = 0 \quad (i \neq j), \quad w_{ij} = 0 \quad (i,\,j = 2,\,3). \tag{5.9}$$

Let us now suppose that the components $s_q^{(1)}$ and $s_q^{(2)}$ do not display critical behavior. Following familiar transformations the Hamiltonian (5.5) can be reduced to the well-studied Ising Hamiltonian

$$\mathcal{H}_I = -\frac{1}{2}\int_{q}(r_{33}+q^2)\,s_q^{(3)}s_{-q}^{(3)} - \int_{q}\int_{q_1}\int_{q_2}\bar{u}_{33}s_q^{(3)}s_{q_1}^{(3)}s_{q_2}^{(3)}s_{-q-q_1-q_2}^{(3)}, \tag{5.10}$$

where

$$\bar{u}_{33} = u_{33} - \sum_{i=1}^{2}\frac{(w_{i3})^2}{2r_{ii}^{(1)}}. \tag{5.11}$$

In going over to the Hamiltonian (5.11) we have used the relations

$$r_{33} < r_{11}^{(1)}, \quad r_{33} < r_{22}^{(1)}, \quad r_{13}^{(1)} = r_{23}^{(1)} = w_{23} = r_{12}^{(1)} = 0. \tag{5.12}$$

Hamiltonian (5.11) has an Ising-like fixed point if

$$\bar{u}_{33} > 0. \tag{5.13}$$

Thus, analysis of anisotropic ferromagnetic materials on the basis of the three-component model for $H=0$ shows that the Landau–Ginzburg–Wilson Hamiltonian has two types of fixed points—of the XY and Ising types. Consequently, second-order phase transitions in anisotropic ferromagnetic materials can be described by the critical exponents corresponding to these models, which are given in terms of the ε expansion by the expressions:

$$\alpha = -\frac{(n-4)}{2(n+8)}\varepsilon - \frac{(n+2)}{4(n+8)^2}(n+28)\,\varepsilon^2 + O(\varepsilon^3), \tag{5.14}$$

$$\beta = \frac{1}{2} - \frac{3}{2(n+8)}\varepsilon + \frac{(n+2)(2n+1)}{2(n+8)^2}\varepsilon^2 + O(\varepsilon^3), \tag{5.15}$$

$$\gamma = 1 + \frac{n+2}{2(n+8)^2}\varepsilon + \frac{n+2}{4(n+8)^3}(n^2+22n+52)\,\varepsilon^2 + O(\varepsilon^3), \tag{5.16}$$

$$\delta = 3 + \varepsilon + \frac{1}{2(n+8)}(n^2+14n+60)\,\varepsilon^2 + O(\varepsilon^3); \tag{5.17}$$

here $\varepsilon = 4 - d$, n is the number of spin components (the spin dimensionality), and d is the dimensionality of the lattice.

The three-dimensional Ising and XY models are characterized respectively by the values $n=1$ and $n=2$ ($d=3$), and the values of the critical exponents

corresponding to these models are given in Table III. The values of ω given here were calculated from scaling relation 7. This exponent for the Ising model was calculated directly by Burkhard and Gunton[43] using the ε expansion. They showed that

$$\frac{1}{T_c(H)} = \frac{1}{T_c(0)} - k\frac{\Delta \cdot H^2}{(zJ)^3}, \tag{5.18}$$

where k is the Boltzmann constant, Δ is of order ε, and J is the exchange integral. Analogous results were obtained by Suzuki[48] in the $1/n$ expansion and by Elliot, Pfeuty, and Wood.[49] A similar result was also obtained by Stinchcombe[50] for one-dimensional magnetic systems with ordering of the easy-plane type.

In summary, theoretical calculations based on both molecular field theory and various model representations and also the results of experiments point to the existence of a second-order phase transition of the fluctuational type in anisotropic ferromagnetic materials in weak magnetic fields.

6. NONMAGNETIC PROPERTIES OF ANISOTROPIC FERROMAGNETIC MATERIALS IN WEAK MAGNETIC FIELDS

As we have established in the preceding sections, the magnetic properties of anisotropic ferromagnetic materials in a magnetic field display anomalies characteristic of second-order phase transitions at a certain temperature $T_c(H)$. Inasmuch as the features of the temperature and field dependence of the nonmagnetic properties of ferromagnets are determined by the magnetic state, it is clear that at $T_c(H)$ there should also be anomalies of the thermal, elastic, electrical, galvanomagnetic, optical, and other properties. In particular, one can predict over a wide range of temperatures the constants characterizing the magnetization[52] and the rotation of the plane of polarization of light,[51] which are effects of great interest for practical applications.

For convenience of discussion all the properties of ferromagnetic materials can be divided into three groups:

1. Properties linear in the magnetization.
2. Properties quadratic in the magnetization.
3. Properties proportional to the temperature coefficient of the magnetization.

1. The properties linear in the magnetization include, first of all, the Kerr and Faraday magnetooptic effects. The angle of rotation of the plane of polarization of light passing through a crystal or reflected from its surface is proportional to the magnetization:

$$\varphi_{F,K} = VM, \tag{6.1}$$

TABLE III.

Model	α	β	γ	δ	ω
Ising	0.0772	0.340	1.244	4.46	2.94
XY	−0.02	0.360	1.300	4.46	0.622

where V is the Verdet or Kundt constant for the Faraday or Kerr effect, respectively; this constant is independent of T and H. In the region of the Curie point the magnetization remains constant up to $T_c(H)$ [see formula (2.9)] and

$$\varphi_{F,K} = \frac{VH}{D}. \tag{6.2}$$

It follows from this relation that in weak magnetic fields $\varphi_{F,K}$ depends linearly on H and remains constant to temperatures all the way up to $T = T_c(H)$, since M = const. For $T = T_c(H)$ one observes the sharp decrease in φ_F characteristic of the kink effect. In fact, experimental studies of the ferromagnetic substances $CrBr_3$ and $Y_3Fe_5O_{12}$ carried out by Litster and Ho[51] and our measurements for epitaxial films of $Y_2BiGa_{1.2}Fe_{3.8}O_{12}$ have shown that the angle of rotation of the plane of polarization in the Faraday effect remains constant over a wide range of temperatures and depends linearly on H at all $T < T_c(H)$ (see Fig. 13).

2. Let us consider the features of the properties which are quadratic in the magnetization for the illustrative case of magnetostriction.[52] Let us consider an isotropic, nonuniformly magnetized ferromagnetic material. In weak fields the magnetostriction λ of such a ferromagnetic near T_c is due to a change in the direction of M, and one can therefore neglect effects which are due to a change in the absolute value of M. In this approximation the magnetoelastic energy near T_c can be written in the form[53]

$$F_{N,U} = \lambda_{xx}\sigma_{xx}M^2 \ (N, U); \tag{6.3}$$

here the subscripts N and U refer to the nonuniformly and uniformly magnetized phases, respectively, M(N) and M(U) are given by expressions (2.8) and (2.9), and λ_{xx} and σ_{xx} are the components of the magnetostriction and elastic strain tensors.

When the magnetoelastic energy is taken into account the phase transition from the nonuniformly magnetized state to the uniform state will occurr only in the case $F_N = F_U$. It then follows from (6.3) and (2.9) that

$$\lambda = -\frac{\partial F_N}{\partial \sigma_{xx}} = -\frac{\partial F_U}{\partial \sigma_{xx}}, \quad \lambda_N = \lambda_U, \tag{6.4}$$

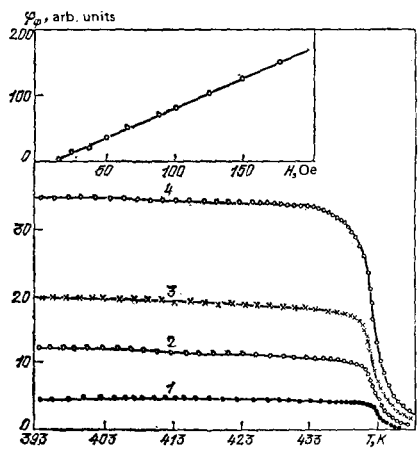

FIG. 13. Dependence of φ_F on T and H for $Y_2BiGa_{1.2}Fe_{3.8}O_{12}$. 1) 17 Oe, 2) 25 Oe, 3) 37 Oe, 4) 55 Oe.

where λ_N and λ_U are the values of the magnetostriction of the nonuniform and uniform phases at $T=T_c(H)$. In our quadratic (in the magnetization) approximation the magnetoelastic energy λ is given by the expressions

$$\lambda_N=\lambda_{xx}\left(\frac{H}{D}\right)^2, \tag{6.5}$$

$$\lambda_U=\sigma_{xx}M_U^2 \quad (s=1/2) \tag{6.6}$$

for the N and U phases, respectively [where M_U is given by formula (2.7)].

Thus it follows from formulas (6.4)–(6.6) that for a ferromagnetic sample with demagnetizing factor D the electrostriction λ has the following features in weak fields: 1) in the nonuniformly magnetized phase λ is independent of T and depends quadratically on H, 2) the values of λ corresponding to $T_c(H)$ determine the temperature dependence of the spontaneous magnetostriction near $T_c(H)$. It should be noted that these features are also present in uniaxial ferromagnetic materials.

We have made an experimental check on the indicated behavioral features in polycrystalline nickel (99.99% pure) and in yttrium and gadolinium ferrite garnets. In the vicinity of the Curie point a capacitive dilatometer with a sensitivity not worse than 10^{-8} was used to record over 50 isotherms for each sample. The measurements in fields up to 500 Oe confirmed the presence of kink phenomena for the magnetostriction. As is seen in Fig. 14, which shows the curves of λ versus the temperature in different fields, the magnetostriction in the temperature region $T>T_c(H)$ remains constant to within the experimental error of 1%. For $T=T_c(H)$ a kink appears on the $\lambda=f(\tau)$ curve, and the kink temperature shifts to lower temperatures with increasing H in accordance with (2.10). As is seen in Fig. 15, in the temperature range $T<T_c(H)$ the experimental points conform well to the straight line representing a dependence of the form $\lambda=f(H^2)$. It should be noted that the kink effect in the magnetostriction can be used to determine the temperature dependence of the spontaneous magnetostriction.[52]

3. Specific heat and magnetocaloric effect. The effect that is perhaps the most thoroughly studied from both the theoretical[6,10,11] and experimental[3,41] standpoint is the influence of a weak magnetic field on the behavior of the specific heat near T_c. Already in the first experimental study of van der Hoeven, Teaney, and

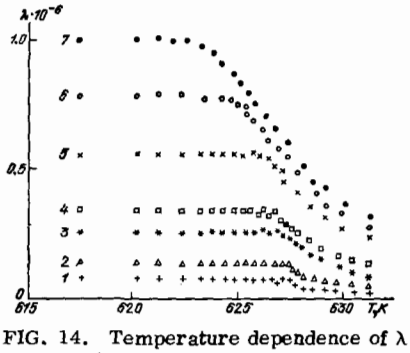

FIG. 14. Temperature dependence of λ for nickel in various fields. 1) 60 Oe, 2) 150 Oe, 3) 200 Oe, 4) 250 Oe, 5) 300 Oe, 6) 350 Oe, 7) 400 Oe.

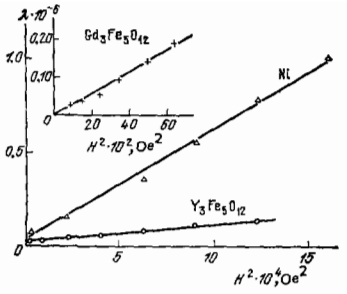

FIG. 15. Dependence of λ on H^2 near the Curie point.

Moruzzi,[3] who investigated the influence of H on the specific heat of the isotropic ferromagnetic substance EuS, it was shown that the peak of the specific heat is shifted to lower temperatures with increasing H. This behavior of the specific heat c_p was explained by Arrott[4] and Wojtowicz and Rayl[6] in the molecular-field approximation for the model of a thin toroid. According to the ideas developed in these papers, the specific heat of a nonuniformly magnetized ferromagnet in the N and U phases is given by the expressions

$$C_U=C_0+\frac{(1-\sigma_U^2)(h-d\sigma_U+\sigma_U)^2}{\tau^2-\tau(1-\sigma_U^2)(1-d)}, \tag{6.7}$$

$$C_N=C_0+\frac{(1-\sigma_N^2)\sigma_N^2}{\tau^2-\tau(1-\sigma_N^2)}, \tag{6.8}$$

from which it follows that there is a discontinuity in the specific heat at $T=T_c(H)$. As H increases, the discontinuity Δc_p shifts to lower temperatures and decreases in absolute value. For $h/d\approx 1$ there is a broad peak above T_c, and the discontinuity disappears (see Fig. 16). These features of the temperature dependence of the specific heat in weak magnetic fields have been confirmed for anisotropic ferromagnetic materials by calculations based on the Landau theory of second-order phase transitions. In particular, the theory predicts that the specific heat at T_c will suffer a discontinuity given by the expression

$$\Delta c_p=\frac{1}{2b}\left[\tau_0(h)-\left(\frac{h_X}{2k_1}\right)^2\right]. \tag{6.9}$$

The characteristic specific-heat anomalies in the temperature dependence have been confirmed by experimental studies.[3,54] Figure 17 shows the $c_p(T)$ curves for Gd in various magnetic fields. The experimental data indicate that there are anomalies in c_p and $T_c(H)$, but the discontinuity predicted by mean field theory does not appear. The anomaly (but not a discontinuity)

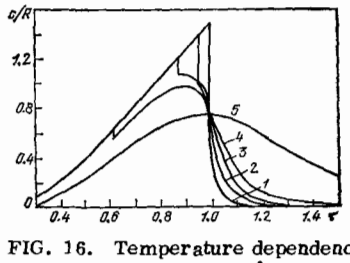

FIG. 16. Temperature dependence of the specific heat in various magnetic fields.[6] 1) $h/d=0.2$, 2) 0.4, 3) 0.6, 4) 0.9, 5) 5.0 ($d=0.04$).

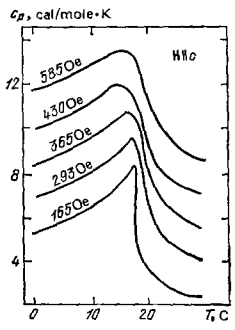

FIG. 17. Temperature dependence of the specific heat of Gd in weak magnetic fields.[54]

occurring at $T_c(H)$ should be attributed to the circumstance that fluctuations in the perpendicular (to the field) component of the magnetization exert a large influence on the temperature dependence of c_p, or to an instability of the nonuniformly magnetized mode.

The magnetocaloric effect in weak magnetic fields has been examined only by Nikitin and co-workers,[37,38] who showed that in easy-plane ferromagnetic materials the ΔT effect grows in size with increasing field and remains negative. Starting at a certain critical field corresponding to a second-order phase transition, a positive component of the magnetocaloric effect appears. These features of the magnetocaloric effect are confirmed by the experimental data (Fig. 18) for the alloys Tb_xCd_{1-x}, which are ferromagnetic substances whose axis of hard magnetization is along the hexagonal axis.

7. FEATURES OF CERTAIN DYNAMIC CRITICAL PHENOMENA IN WEAK MAGNETIC FIELDS

The critical dynamics of ferromagnetic materials in weak magnetic fields was studied in Refs. 55–68.

Because the critical dynamics of the spin systems of ferromagnetic materials is most often studied in experiments on the propagation of ultrasonic waves and on the susceptibility in alternating magnetic fields (the dynamic susceptibility), let us briefly discuss the features of these dynamical parameters in anisotropic ferromagnetic materials at $H \neq 0$.

It is known from experimental and theoretical studies[56-61] that magnetically ordered crystals exhibit an ultrasonic absorption peak and velocity minimum near T_c for $H = 0$. It has been established that these anomalies are due to the presence of a spin-phonon interaction of a magnetostrictive nature. The spin-phonon interaction responsible for these critical anomalies arises on account of the modulation of the exchange interaction by elastic strains. As a result, the spin fluctuations give rise to random forces which affect the normal acoustic modes and lead to an anomalous attentuation of elastic waves. According to the theory of Mori,[60] the absorption coefficient α_K and the relative change in the speed $\Delta v/v$ are expressed in terms of the temporal correlation function of the random forces and are proportional to the four-spin correlation function, which carries all the dynamical information about the spin system.

A magnetic field suppresses the spin fluctuations and removes the critical anomalies. However, the suppression of fluctuations is not observed in pure form in all ferromagnetic materials. In some materials α_K increases in a magnetic field,[36,62,63] while in other materials it decreases.[36,61] To explain the different nature of the effect of H on α_K, a polarization mechanism has been proposed for the anomalous absorption.[36] With allowance for this mechanism, the absorption coefficient α_K in a magnetic field $H \| z$ is given by the expression[36]

$$\alpha_K = (2\rho V v_l)^{-1} \operatorname{Re} \Big[4 g_0^{z*}(k)\, g_0^z(k)\, \langle s_0^z \rangle \int_0^\infty (s_K^z(t),\ s_K^z(0)) \exp(-i\omega_K t)\, dt \qquad (7.1)$$

$$+ \sum g_q^{\alpha *}(k)\, g_{q'}^{\alpha'}(k) \int_0^\infty (s_q^\alpha(t)\, s_{-q-K}^{\alpha'}(t),\ s_{-q'}^{\alpha'}(0)\, s_{q'+K}^{\alpha'}(0)) \exp(-i\omega_K t)\, dt \Big],$$

$$g_q^\alpha(k) = \sum_j \exp(iq R_{ij}) [\exp(ikR_{ij}) - 1] e_K \frac{\partial J_{ij}}{\partial R_i}, \qquad (7.2)$$

where ρ and V are the density and volume of the crystal, v_l is the speed of propagation of longitudinal waves, e_K and ω_K are the phonon polarization vector and frequency, J_{ij} is the exchange integral, R_i is the position vector of the i-th lattice site, and $\langle s_0^\alpha \rangle$ is the static spin polarization. The first term in this expression, consisting of the product of the spin polarization and the two-spin correlation function, leads to an increase in the absorption coefficient in the limit of weak magnetic fields. The second term, which is governed by the four-spin correlation function, causes a decrease in α_K on account of the suppression of fluctuations by the magnetic field.

There are thus two competing mechanisms for the anomalous absorption of ultrasonic waves in a magnetic field, and it can turn out that their contributions to α_K are equal at a certain temperature—this so-called compensation temperature is determined by the range of the exchange interaction. Below the compensation temperature the suppression of critical fluctuations by the magnetic field is the dominant mechanism, and the absorption coefficient α_K decreases with H. Above this temperature α_K is determined by the spin polarization, and a magnetic field therefore leads to an increase in α_K.

In anisotropic ferromagnetic materials the presence of second-order phase transitions induced by the mag-

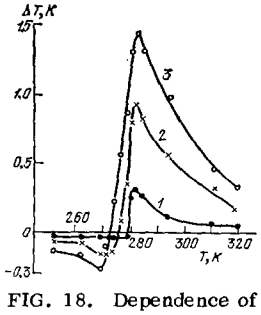

FIG. 18. Dependence of the magnetocaloric effect on T and H for the alloy $Tb_{0.2}Gd_{0.8}$ (Ref. 38). 1) 5.0 kOe, 2) 9.1 kOe, 3) 12 kOe.

netic field leads to interesting features in the field dependence and temperature dependence of the absorption coefficient and speed of ultrasonic waves. It has been shown experimentally in MnP that two ultrasonic absorption peaks appear in a magnetic field perpendicular to the easy axis (the c axis). One of them is located just above $T_c(0)$ and is shifted to higher temperatures with increasing H. The other absorption peak is observed below $T_c(0)$ and is due to the ferromagnetic-paramagnetic transition. This absorption peak is shifted to lower temperatures with increasing H (Fig. 19). The presence of a second-order phase transition in MnP for $\mathbf{H} \perp c$ is also indicated by the data on the field dependence of α_K. If the temperature is fixed at a value below $T_c(0)$ and the external magnetic field is varied, the absorption coefficient will pass through a maximum at a certain value $H = H_{cr}$. As the temperature is decreased further below $T_c(0)$, this peak shifts toward higher fields, qualitatively confirming the H-T diagram for uniaxial ferromagnetic materials. Analogous features in the temperature and field dependence of the acoustic absorption have been observed by the present authors in gadolinium (see Fig. 20), and for the speed of sound by Jiles and Palmer.[64]

It follows from (7.1) that for a quantitative description of the features of α_K and $\Delta v/v$ it is necessary to calculate the temperature and field dependence of the two- and four-spin correlation functions. For calculating the four-spin correlation function one can use the following decoupling, which enables one to express this function in terms of the two-spin correlators:

$$(ab,\ cd) \sim (a, b)(c, d) + (a, c)(b, d) + (a, d)(b, c) - \langle a, b\rangle\langle c, d\rangle. \tag{7.3}$$

This decoupling is valid as long as the correlation length is smaller than the range of the exchange interaction. In magnetic metals, where the exchange interaction is of a long-range character, the decoupling (7.3) works well over the entire temperature range except very close to $T_c(H)$. Applying (7.3) to (7.1), using the familiar relation between the two-spin correlation functions and the susceptibility

$$(s_q^\alpha,\ s_{-q}^\alpha) = (g\mu_\sigma)^{-2}\chi_q^\alpha, \tag{7.4}$$

one can transform the expression for α_K for, say, an orthorhombic crystal ($\mathbf{H} \| b$ axis, $\omega_K \tau_K \ll 1$, where τ_K is the relaxation time) to the form

$$\alpha_K^b = F_K\tau\left\{\left(\frac{\sigma(1-b)}{\tau-(1-k_b)(1-b)}\right)^2 + G\tau\left[\left(\frac{1-b}{\tau-(1-k_b)(1-b)}\right)^{3/2} + \left(\frac{1-b/3}{\tau-(1-k_a)(1-b/3)}\right)^{3/2} + \left(\frac{1-b/3}{\tau-1+b/3}\right)^{3/2}\right]\right\}. \tag{7.5}$$

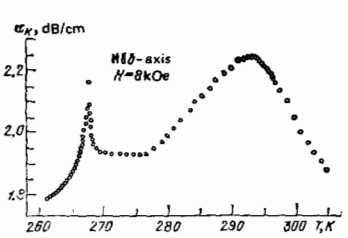

FIG. 19. Temperature dependence[63] of α_K in MnP.

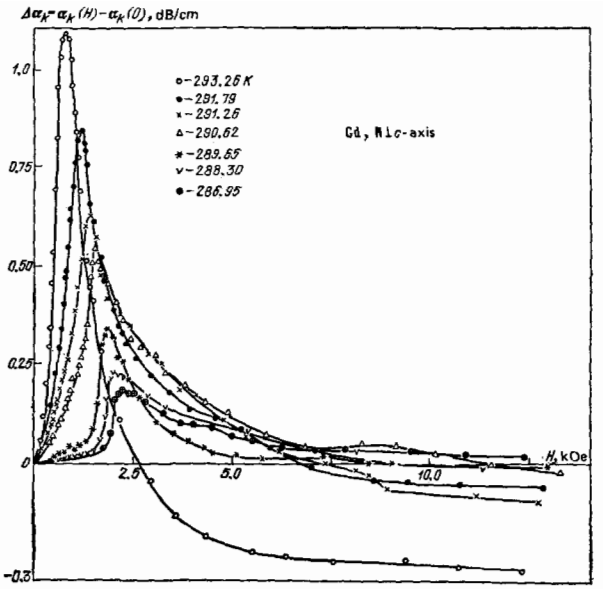

FIG. 20. Field dependence of $\Delta\alpha_K$ in Gd near T_c.

In an anlogous way one can obtain an expression for the change in the speed of sound:

$$\Delta v = -F_K\frac{3B}{|s(s+1)k^2}\left\{\sigma^2\left(\frac{1-b}{\tau-1+b}\right) + 8G\tau\left[\sqrt{\frac{1-b}{\tau-1+b}} + \sqrt{\frac{1-(b/3)}{\tau-(1-k_a)(1-(b/3))}} + \sqrt{\frac{1-(b/3)}{\tau-(1-k_b)(1-(b/3))}}\right]\right\}; \tag{7.6}$$

here $k_i = (J_0^c - J_0^i)/J_0^c$ is the anisotropy constant ($i = a, b$), F_K and G are constants characterizing the magnetoelasticity and the range of the exchange interaction, respectively, B is a constant characterizing the material, k is the wave number, σ is the spin polarization normalized by s, b is defined by Eq. (46) of Ref. 36, and the remaining notation is as before. These expressions permit a quantitative description of the experimentally observed features of the temperature and field dependence of α_K and $\Delta v/v$. In particular it follows from (7.5) that the low-temperature peak in α_K (see Fig. 19) is due to the fourth term, which goes to infinity at $T = T_c[1 - (b/3)]$, and the broad peak above $T_c(0)$ is due to the first and second terms. The physical cause of the two anomalies in α_K, as follows from (7.5), is the interaction of the sound waves with fluctuations of the spin components along the c and b axes, the correlation lengths of which, according to the calculations of Sznajd,[10] pass through maxima at $T = T_c(H)$ and $T_c(0)$. In a similar way one can also explain the anomalies in the speed of longitudinal sound waves in Gd on the basis of (7.6).

Let us now consider the features in the behavior of the dynamic susceptibility at temperatures near the second-order phase transition observed in a weak magnetic field. The dynamic susceptibility as we know, is a complex quantity:

$$\chi = \chi' + i\chi''. \tag{7.7}$$

When $\omega\tau_K \ll 1$ the real part χ' of the susceptibility is approximately equal to the static susceptibility, which implies that χ' in the ferromagnetic phase does not depend on the temperature. At $T = T_c(H)$ the compo-

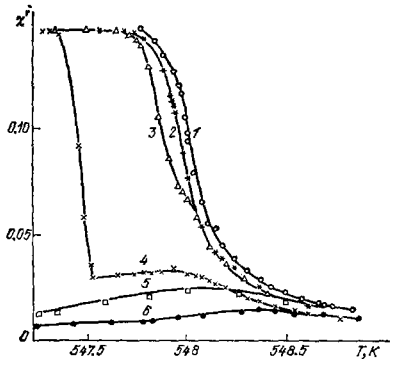

FIG. 21. Temperature dependence[67] of χ' in an external magnetic field for a spherical sample of $Y_3Fe_5O_{12}$ at a frequency of 1 MHz. 1) 0, 2) 3.4 Oe, 3) 8 Oe, 4) 40 Oe, 5) 60 Oe, 6) 128 Oe.

nent χ' begins to decrease sharply, and above $T_c(0)$, according to (3.17), it passes through a maximum whose position is shifted to higher temperatures with increasing H. Behavior of this nature for the temperature dependence of χ' has been observed experimentally[65-67] in $Y_3Fe_5O_{12}$ (Fig. 21).

The imaginary component χ'' of the susceptibility, which characterizes the power absorbed by the spin system from an rf field, is proportional to the absorption coefficient of longitudinal sound waves.[36] In fact, the propagation of a sound wave is accompanied by dissipation of energy in the spin system:

$$P=\alpha_K V\rho\omega_K^3 u_0^2 v_l, \tag{7.8}$$

where u_0 is the atomic displacement caused by the sound wave. The dissipation of the rf-wave energy, on the other hand, is determined by the imaginary component of the susceptibility.

$$P=\frac{\omega_K}{2}\chi''H_\sim^2. \tag{7.9}$$

It follows from (7.8) and (7.9) that χ'' is proportional to α_K, and in a magnetic field perpendicular to the anisotropy axis the $\chi''(T)$ curve should display two peaks, corresponding to the temperatures $T_c(H)$ and $T_c(0)$. Figure 22 shows the experimental data for a spherical sample of $Y_3Fe_5O_{12}$, confirming the presence of two peaks. The dynamics of these peaks in a static magnetic field is the same as that of α_K.

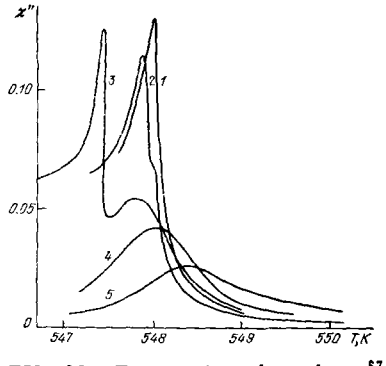

FIG. 22. Temperature dependence[67] of χ'' in an external magnetic field for a spherical sample of $Y_3Fe_5O_{12}$ at a frequency of 1 MHz. 1) 0, 2) 8 Oe, 3) 40 Oe, 4) 60 Oe, 5) 128 Oe.

In summary, studies of the propagation of ultrasonic waves and the dynamic susceptibility confirm the presence of second-order phase transitions in weak magnetic fields in anisotropic or nonuniformly magnetized ferromagnetic materials. Furthermore, these experiments yield the most correct reconstruction of the H-T phase diagram, since α_K and χ'' pass through clearly defined peaks at $T=T_c(H)$ (see Figs. 20 and 22), and for this reason the error in determining H_{cr} or $T_c(H)$ is significantly smaller than for magnetic and other types of measurements.

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