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Phase diagrams near the Lifshitz point. III. Tetragonal crystals with an easy plane of magnetization

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Phase diagrams near a Lifshitz point are considered for tetragonal crystals with an easy plane of magnetization. The modulated phase is shown to be sinusoidal or helicoidal, depending on whether a certain parameter μ measuring the strength of tetragonal anisotropy is larger or smaller than 1. The line $T_H(P)$ of first-order phase transitions between the ferromagnetic and modulated phases is shown to be tangent to the order-disorder transition line $T_\lambda(P)$ at the Lifshitz point. For $\mu < 1$, the line $T_H(P)$ is shown to be displaced in the direction of the helicoidal phase with the growth of μ. Such displacement of this line with the increase of crystal anisotropy is shown to be a general property of phase diagrams in the vicinity of the Lifshitz point.

I. INTRODUCTION

The concept of Lifshitz point was introduced by Hornreich *et al.*[1,2] A Lifshitz point (P_L, T_L) is a triple point on the P-T diagram (T is temperature, and P is some other thermodynamic parameter, like pressure or material composition) between the paramagnetic, ferromagnetic, and modulated (helicoidal or sinusoidal) phases; a characteristic feature of the Lifshitz point is that the modulation wave vector $\vec{k}_0$ increases *continuously* from zero as one moves from (P_L, T_L) into the modulated phase. Hornreich *et al.*[1,2] found the Lifshitz point to be a multicritical point and obtained the relevant critical exponents and scaling relations, using renormalization-group techniques. They also suggested that Lifshitz points might occur in various magnetic compounds and mixtures. The results of Hornreich *et al.* stimulated further theoretical work on Lifshitz points and related phenomena.[3-5] One of the subjects of interest is the thermodynamic behavior of a system in the vicinity of a Lifshitz point and the shape of the phase diagram in this vicinity.

In our previous papers,[5] (hereafter referred to as I and II) we considered phase diagrams near (P_L, T_L) for uniaxial magnetic systems and for systems with cylindrical and hexagonal (or rhombohedral) symmetry having an easy plane of magnetization. We found that the thermodynamic properties near the Lifshitz point depend on the symmetry of the paramagnetic phase (phase I) of the system. The results of Papers I and II concerning this dependence are as follows:

In the case of cylindrical symmetry, the phase transitions between the ferromagnetic phase (phase II) and the modulated phase (phase III) are second order, and the line $T_H(P)$ of these transitions meets the line $T_\lambda(P)$ of order-disorder transitions under some angle. The presence of hexagonal anisotropy makes the II $\rightleftharpoons$ III phase transitions first order, and displaces the line $T_H(P)$ into the region occupied in the cylindrical case by phase III. (This region is determined by the inequality $\alpha(P, T) < 0$, where α is a coefficient in the expansion of the free energy [see I and II, Eq. (2.3)].) The lines $T_H(P)$ and $T_\lambda(P)$ remain nontangent at their meeting point (P_L, T_L). Finally, in the uniaxial case the line $T_H(P)$ becomes tangent to the line $T_\lambda(P)$ at (P_L, T_L), which means a still deeper "penetration" of the ferromagnetic phase into the region of $\alpha < 0$. Since a uniaxial system may be regarded as the most anisotropic of the three types of systems considered, the above results suggest that the stronger the crystal anisotropy is, the larger is the area of the phase diagram which the ferromagnetic phase "captures" from the modulated phase. Systems with tetragonal symmetry having an easy plane of magnetization occupy—in the sense of anisotropy—an intermediate position between uniaxial systems and hexagonal systems with an easy plane. Therefore it is of interest to study the phase diagram associated with a Lifshitz point for such systems and to compare the results with those obtained in Papers I and II. Such a study is undertaken in the present paper.

The formalism used in this paper is essentially the same as in Paper II. The ordered phases are characterized by a two-component order parameter $\vec{M} = M_x \hat{x} + M_y \hat{y}$ representing the magnetization in the easy plane. The possible spatial variation of $\vec{M}$ is restricted to the z direction (parallel to the tetragonal axis). The free-energy functional $F(\vec{M})$ is expanded in terms of $\vec{M}$ and its first- and second-order derivatives with respect to z. The results of this work are based on the minimization of $F(\vec{M})$; therefore they are essentially mean-field

results, as in Papers I and II.

II. STUDY OF THE PHASE DIAGRAM

In a tetragonal crystal with an easy plane of magnetization, the expansion of $F(\vec{M})$ has the form

$$F(\vec{M}) = \int \{\tfrac{1}{2}A_0(M_x^2+M_y^2)+\tfrac{1}{4}B(M_x^2+M_y^2)^2 + \tfrac{1}{4}B_1 M_x^2 M_y^2 + \tfrac{1}{2}\alpha[(M_x')^2+(M_y')^2] + \tfrac{1}{4}\beta[(M_x'')^2+(M_y'')^2]\}\, d^3r , \quad (2.1)$$

where the primes denote differentiation with respect to z. From the requirement for thermodynamic stability it follows that

$$B>0 , \quad B+\tfrac{1}{4}B_1>0 , \quad \beta>0 . \quad (2.2)$$

The Lifshitz point (P_L, T_L) is the intersection point of the lines

$$A_0(P,T)=0 \quad (2.3)$$

and

$$\alpha(P,T)=0 . \quad (2.4)$$

The order-disorder transition line $T_\lambda(P)$ is given by the same equations as in the cases considered in Papers I and II. The part of this line lying in the region $\alpha>0$ coincides with the line $T_0(P)$ defined by Eq. (2.3); this part corresponds to second-order transitions between the paramagnetic phase (phase I) and the ferromagnetic phase (phase II). The part of $T_\lambda(P)$ lying in the region $\alpha<0$ is described by the equation

$$A_{k_0}=A_0-\alpha^2/2\beta=0 . \quad (2.5)$$

This part corresponds to second-order transitions between the paramagnetic phase and the modulated phase (phase III). The two parts of $T_\lambda(P)$ have a common tangent at (P_L, T_L).

Let us consider the ferromagnetic phase (phase II). The vector of spontaneous magnetization in this phase $\vec{M}_{II}$, which is determined by the minimization of $F(\vec{M})$, with $M_x'=M_y'=0$, depends on the sign of B_1. If $B_1>0$, then

$$\vec{M}_{II}=\pm M_{II}\hat{x} \quad \text{or} \quad \vec{M}_{II}=\pm M_{II}\hat{y} , \quad (2.6)$$

where

$$M_{II}=(-A_0/B)^{1/2} . \quad (2.7)$$

Accordingly, the free energy at thermodynamic equilibrium in phase II is

$$F_{II}=-A_0^2/4B . \quad (2.8)$$

If $B_1<0$, then the rotation of the coordinate system about the z axis by the angle $\frac{1}{4}\pi$ yields

$$\vec{M}=M_\xi\hat{\xi}+M_\eta\hat{\eta} , \quad (2.9)$$

where

$$\hat{\xi}=(1/\sqrt{2})(\hat{x}-\hat{y}) , \quad \hat{\eta}=(1/\sqrt{2})(\hat{x}+\hat{y}) , \quad (2.10)$$

$$M_x=(1/\sqrt{2})(M_\xi+M_\eta) , \quad M_y=(1/\sqrt{2})(M_\eta-M_\xi) ; \quad (2.11)$$

so that

$$F(\vec{M}) = \int \{\tfrac{1}{2}A_0(M_\xi^2+M_\eta^2)+\tfrac{1}{4}b(M_\xi^2+M_\eta^2)^2 + \tfrac{1}{4}b_1 M_\xi^2 M_\eta^2 + \tfrac{1}{2}\alpha[(M_\xi')^2+(M_\eta')^2] + \tfrac{1}{4}\beta[(M_\xi'')^2+(M_\eta'')^2]\}\, d^3r , \quad (2.12)$$

where

$$b\equiv B+\tfrac{1}{4}B_1>0 , \quad b_1\equiv -B_1>0 . \quad (2.13)$$

We see that in the new coordinates, ξ and η, the situation reduces to the previous one. We shall therefore consider only the case of $B_1>0$.

Let us now consider the modulated phase (phase III). In this phase, the magnetization at thermodynamic equilibrium, $\vec{M}_{III}$, is a function of z. In order to find $\vec{M}_{III}(z)$, one has to solve the variational equations

$$\tfrac{1}{2}\beta M_x''''-\alpha M_x''+A_0M_x+B(M_x^2+M_y^2)M_x+\tfrac{1}{2}B_1M_y^2M_x=0 , \quad (2.14)$$

$$\tfrac{1}{2}\beta M_y''''-\alpha M_y''+A_0M_y+B(M_x^2+M_y^2)M_y+\tfrac{1}{2}B_1M_x^2M_y=0 .$$

As in the uniaxial case, the solutions of these equations cannot be obtained in an analytical form. However, it is known (see I, Appendix A) that at $T\to T_\lambda(P)-0$, $\vec{M}_{III}(z)$ has the asymptotic form

$$\vec{M}_{III}\approx M_{xk_0}\cos(k_0z+\phi)+M_{yk_0}\sin(k_0z+\psi) , \quad (2.15)$$

where $k_0=(-\alpha/\beta)^{1/2}$. The parameters M_{xk_0}, M_{yk_0}, and $\psi-\phi$ must be found from the minimization of the expression $F(M_{xk_0}, M_{yk_0}, \psi-\phi)$ obtained by substituting (2.15) for $\vec{M}$ in $F(\vec{M})$. This expression is

$$F(M_{xk_0}, M_{yk_0}, \psi-\phi) = \tfrac{1}{4}A_{k_0}(M_{xk_0}^2+M_{yk_0}^2)+\tfrac{3}{32}B(M_{xk_0}^4+M_{yk_0}^4) + \tfrac{1}{16}(B+\tfrac{1}{2}B_1)[2+\cos2(\psi-\phi)]M_{xk_0}^2M_{yk_0}^2 , \quad (2.16)$$

where $A_{k_0}=A_0-\alpha^2/\beta$. The minimization of (2.16) with respect to $\psi-\phi$ yields

$$\psi-\phi=\tfrac{1}{2}\pi+n\pi , \quad n=0,1,\ldots . \quad (2.17)$$

Substituting (2.17) into (2.16), we have

$$F(M_{xk_0}, M_{yk_0})=\tfrac{1}{4}A_{k_0}(M_{xk_0}^2+M_{yk_0}^2)+\tfrac{3}{32}B(M_{xk_0}^2+M_{yk_0}^2)^2 + \tfrac{1}{32}(B_1-4B)M_{xk_0}^2M_{yk_0}^2 . \quad (2.18)$$

Further results depend on whether $\mu>1$ or $\mu<1$, where $\mu\equiv B_1/4B$. If $\mu>1$, the minimization of

$F(M_{xk_0}, M_{yk_0})$ yields

$$M_{yk_0}=0\,,\quad M_{xk_0}\equiv 2M_{k_0}=2(-A_{k_0}/3B)^{1/2} \tag{2.19}$$

or vice versa; so that

$$\vec{M}_{III}=2M_{k_0}\cos(k_0z+\phi)\hat{x} \tag{2.20a}$$

or

$$\vec{M}_{III}=2M_{k_0}\cos(k_0z+\phi)\hat{y}\,. \tag{2.20b}$$

It follows that in the case of $\mu>1$ phase III is characterized by a static *transverse sinusoidal wave* of magnetization. The free energy at thermodynamic equilibrium in phase III is in this case

$$F_{III}=-A_{k_0}^2/6B\,. \tag{2.21}$$

According to (2.8), (2.21), the expressions of F_{II} and F_{III} in terms of the expansion coefficients A_0, α, β, B are the same as in the uniaxial case.[6] It follows that the line $T_H(P)$ of first-order transitions between phases II and III and the latent heat ΔQ of these transitions are also described by the same equations as in the uniaxial case [See I, Eqs. (3.11), (3.15), and (3.16)]. In particular,

$$T_H(P)=T_0(P)-2.2\gamma\,(P-P_L)^2\,, \tag{2.22}$$

where the coefficient γ is defined as

$$\gamma\equiv\left[\left(\frac{\partial\alpha}{\partial T}\right)_{P,\,T=P_L,\,T_L}\left(\frac{dT_0}{dP}\right)_{P=P_L}+\left(\frac{\partial\alpha}{\partial P}\right)_{P,\,T=P_L,\,T_L}\right]^2\beta^{-1}C\,, \tag{2.23}$$

with

$$C\equiv\left(\frac{\partial A_0}{\partial T}\right)_{P,\,T=P_L,\,T_L} \tag{2.24}$$

being the Curie-Weiss constant for the magnetic susceptibility in the paramagnetic phase near the point (P_L, T_L).

If $\mu<1$, then the minimization of the expression (2.18) gives

$$M_{xk_0}^2=M_{yk_0}^2\equiv 4M_{k_0}^2=-A_{k_0}/(B+\tfrac{1}{8}B_1)\,, \tag{2.25}$$

so that

$$\vec{M}_{III}=2M_{k_0}[\cos(k_0z+\phi)\hat{x}\pm\sin(k_0z+\phi)\hat{y}]\,. \tag{2.26}$$

It follows that in the case of $\mu<1$, phase III is characterized by a *helicoidal* magnetic structure. The expression for F_{III} is

$$F_{III}=-A_{k_0}^2/(B+\tfrac{1}{8}B_1)\,. \tag{2.27}$$

From the equation

$$F_{II}=F_{III} \tag{2.28}$$

one obtains the equation of the line $T_H(P)$ of first-order transitions between phases II and III:

$$-A_0=[(4+2\mu)^{1/2}-2]^{-1}\alpha^2/\beta\,. \tag{2.29}$$

It follows from (2.29), (2.23), and (2.24) that in the vicinity of the Lifshitz point (P_L, T_L)

$$T_H(P)\approx T_0(P)-[(4+2\mu)^{1/2}-2]^{-1}\gamma(P-P_L)^2\,. \tag{2.30}$$

In the same fashion as in Paper I, Appendix A, it can be shown that the single-harmonic approximation (2.15) is satisfactory throughout phase III in both of the cases considered; the third and higher odd harmonics of k_0—appearing in $\vec{M}_{III}(z)$ because of the terms $B_1M_y^2M_x, B_1M_x^2M_y$ in Eqs. (2.14)—are negligible.

Let us now discuss the results. The term $B_1M_x^2M_y^2$ in the expansion (2.1) characterizes the tetragonal anisotropy, and the parameter $\mu=B_1/4B$ can serve as a measure of the strength of this anisotropy. We see that certain properties of the phase diagram depend on the value of μ. If $\mu>1$ (strong anisotropy), then phase III is sinusoidal, as in the uniaxial case, and the line $T_H(P)$ is tangent to the line $T_0(P)$ [or $T_\lambda(P)$] at the Lifshitz point and does not depend on μ. If $\mu<1$ (weak anisotropy), then phase III is helicoidal, as in the cases of cylindrical and hexagonal symmetry. However, as distinct from these cases, the line $T_H(P)$ is tangent to $T_0(P)$, so that phase II extends deeper into the region of $\alpha<0$ than in these cases. For $\mu<1$, the line $T_H(P)$ depends on μ. As μ increases, approaching the value $\mu=1$, the

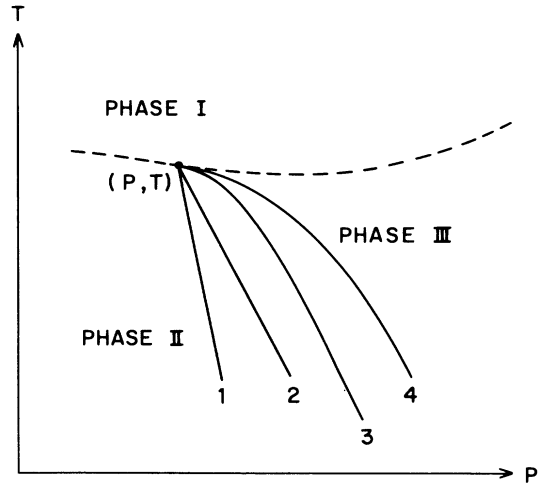

FIG. 1. Relative positions of the II≒III phase-transition line $T_H(P)$ for different types of systems. The positions 1–4 of this line correspond to the following cases: 1—easy plane of magnetization with cylindrical symmetry; 2—easy plane with hexagonal symmetry; 3—easy plane with tetragonal symmetry and $\mu<1$ (weak anisotropy); 4—the same with $\mu>1$ (strong anisotropy). The dashed line is the order-disorder transition line $T_\lambda(P)$.

line $T_H(P)$ is displaced deeper into the region of $\alpha < 0$, approaching its limiting position corresponding to the case of $\mu \geq 1$.

III. CONCLUSION

In conclusion, the results of Papers I, II, and the present one exhibit the existence of the following regularity.

The increase of crystal anisotropy displaces the ferromagnetic-helicoidal phase transition line $T_H(P)$ in the vicinity of the Lifshitz point in the direction of the helicoidal phase—so that the area on the P-T diagram occupied by the ferromagnetic phase increases, and the area occupied by the helicoidal phase decreases—until the helicoidal phase changes into a sinusoidal one. This regularity is quite natural: the ferromagnetic and sinusoidal states are characterized by a certain preferred direction of magnetization, whereas in the helicoidal phase all directions (within the easy plane) are equivalent. The crystal anisotropy tends to align the magnetic moments along a preferred crystallographic direction (easy direction). Therefore the stronger the anisotropy is, the more advantageous the ferromagnetic and sinusoidal states are, as compared with the helicoidal one. This tendency is displayed in the above-mentioned displacement of the line $T_H(P)$ [and in the change of the form of $T_H(P)$ when the hexagonal symmetry changes for a tetragonal, more anisotropic one]. It is also displayed in the eventual replacement of the helicoidal state by the sinusoidal one when the tetragonal anisotropy is sufficiently strong.

The relative positions of the line $T_H(P)$ for the different cases considered are shown schematically in Fig. 1.

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[6]See Eqs. (3.7) and (3.8) in I.