PERIODIC MAGNETIC STRUCTURES AND PHASE TRANSITIONS

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It is proved that a periodic dependence of the magnetic moment on the coordinates can arise in conductors if the temperature is low enough and in a definite range of the magnetic induction B, $2\pi^2 T \lesssim \hbar\Omega \lesssim (4\pi)^{2/3}\epsilon_0(v/c)^{4/3}$ ($\Omega \propto B$ is the cyclotron frequency, ϵ_0 and v are the carrier degeneracy temperature and velocity, and c is the light velocity). The spontaneous ferromagnetic moment may also be a source of the magnetic induction. The periodic structure does not depend on the sample geometry; its amplitude is of the order of $B(v/c)^2(\hbar\Omega/\epsilon_0)^{1/2}$, and its period is of the order of the Larmor radius r. An electric field E, $eE/r \sim \hbar\Omega(\hbar\Omega/\epsilon_0)^{1/2}$, having same period, appears simultaneously. The appearance of the periodic structure is connected with a phase transition of first or second order. In the latter case the phase transition is of Landau type; the relative jump of the heat capacity (if **H** is fixed and the temperature varies) or of the magnetic susceptibility (if T is fixed and **H** varies; in this case the transitions are periodic in H^{-1}) is of the order $(\hbar\Omega/\epsilon_0)^{1/2}$. Near the transition point the oscillations period changes linearly with τ (τ is the deviation of the temperature or of the magnetic field H from the transition point), and the oscillation amplitude is proportional to $\tau^{1/2}$. There exist the isolated critical points (where $c_H \propto \tau^{-2/3}$) and critical points of second-order phase transitions (where the heat capacity c_H or the magnetic susceptibility tends to infinity like $\tau^{-1/2}$).

The singularity near the absolute instability curve of the homogeneous state, where $c_H \propto \tau^{-1/2}$, is determined. The relative "width" (in terms of T or H) of the phase transitions is of the order of $(r/D)^2$, D being the sample characteristic linear dimension of the sample; inside this region all the thermodynamic quantities are analytic functions of T and H, as they should be in a finite sample. The character of the transitions does not depend on the bulk-sample geometry, unlike the transitions to the domain structure (where, for example, a first-order transition takes place if **H** is parallel to the sample surface and of second order if **H** is normal to it). The state diagram is shown at Fig. 1: the number of transition points is on the order of $0.25\,(\epsilon_0/T)(v/c)^{4/3}$ if T is fixed and less than $0.05\,\hbar\Omega$.

1. PHYSICAL CAUSE OF PERIODICITY OF MAGNETIC STRUCTURES AND CHARACTER OF PHASE TRANSITIONS

SHOENBERG[2] has shown (see also[3]) that the field acting on charges in a metal coincides with the magnetic induction B. This means that each charge is acted upon by a magnetic moment produced by all the remaining charges which are located at a distance equal to the Larmor diameter 2r from this charge. As a result, a self-consistent nonlocal interaction is produced between the charges, and leads (Condon[4]) when $4\pi\chi_{max} > 1$ (χ—magnetic susceptibility, the maximum is taken with respect to B) to a stratification into diamagnetic domains (see also[5]).

However, in a system with more than one component, with para- and diamagnetic components, the homogeneous structure of the magnetic moment may turn out to be unstable also when $4\pi\chi_{max} < 1$. As is well known, the oscillating part of the susceptibility is the sum of the susceptibilities produced by all the extremal cross sections of the different carrier bands:

$$\chi = \sum_{\mu=1}^{\mu_0} \chi_\mu.$$

It is possible here that, although the stratification into phases is not favored in a one-component system at such a value of χ (in particular, $4\pi\chi < 1$), instability to stratification would occur already for the paramagnetic components in the absence of an interaction, since $4\pi\chi_0 = 4\pi\sum\chi_\nu > 1$ (ν--those of the μ for which $\chi_\nu > 0$). Consequently, the stratification does not occur precisely as a result of the self-consistent interaction with the components for which the homogeneous state is stable. This means that the interaction leads to a "mixing," i.e., has the character of an effective attraction of the diamagnetic ($\chi < 0$) and paramagnetic ($\chi > 0$) components[2]. Thus, the instability of the components ν gives

[1] A preliminary communication concerning this work was published earlier [1].

[2] The stratification into magnetic phases when $(\partial H/\partial B)_T < 0$ is analogous to the ordinary stratification in a multicomponent system into vapor and liquid when $(\partial p/\partial\rho)_T < 0$; the pressure corresponds to a magnetic-field intensity equal to the sum of the partial intensities for the components corresponding to different extremal sections or zones, and the specific volume $V = \rho^{-1}$ corresponds to magnetic induction. The entire interaction between the components reduces formally to the fact that the "specific volumes" of different components should coincide (B, obviously, depends only on the coordinates!). As a result, although the stratification into phases with B and B′ is convenient for the "components" ν, it is forbidden by the remaining zones, for which such "specific volumes" are not convenient. However, in the case of inhomogeneous induction B, different χ_μ vary differently, the total susceptibility χ can increase, a fact which is thermodynamically convenient (formula (3.9)).

rise to a tendency to stratification into phases, and the attraction between them (negative surface energy) gives rise to mixing. In such a case, it is natural to expect ordering of the phases, i.e., the occurrence of a self-consistent periodic structure, the characteristics of which are determined by the properties of the thermodynamic system. Thus, the period of the structure turns out to be macroscopic and of the order of the "interaction radius"—the Larmor radius r.

It can be understood from general considerations how the transition from the homogeneous structure to the periodic structure can occur[3]. The requirement of thermodynamic stability leads to a continuity of the corresponding thermodynamic potential in the transition (thus, positiveness of the specific heat c_V denotes continuity and monotonicity of the free energy as a function of the temperature T). This means that there can appear at the transition point either an infinitesimally small amount of a new phase with essentially new properties, i.e., finite amplitude A of the spatial oscillations, or else that the new state itself differs infinitesimally little from the old one (i.e., the amplitude of the oscillations is infinitesimally small).

In the former case the production of the new phase is connected with the nuclei that are produced in a fluctuating manner, and in order to be thermodynamically favored these nuclei must therefore be sufficiently large (since there is a surface energy on the boundary of the different phases). Therefore superheating and supercooling--conservation of the metastable homogeneous phase—are possible. The transition point is consequently the point of intersection of the thermodynamic potentials of the homogeneous phases and is not in essence a singular point (see[7], Secs. 81, 83)—this is a first-order phase transition.

In the latter case there is no surface energy, and therefore superheating or supercooling is impossible. The new state appears suddenly in the entire volume, and the transition point is a singular point of the thermodynamic potential of the system. The change of the state immediately in the entire macroscopic volume should, obviously, be "prepared beforehand." The dimension of the fluctuationally arising regions of the unfavored state (the correlation radius r_c) should increase without limit on approaching the transition point. In Landau's theory this occurs in accordance with the law ([7], Sec. 119)

$$r_c \sim a\left|\frac{T-T_c}{T_c}\right|^{1/2} = a\tau^{1/2}$$

(a—distance between particles, T_c—transition temperature). So long as the correlation radius is small compared with the interaction radius ρ (if this is possible), in our case the Larmor radius, the fluctuation inhomogeneity of the oscillation amplitude can be disregarded and this amplitude can be regarded as homogeneous and "reliable" (and not accidental), determined from the thermodynamic-equilibrium conditions. The inequality $r_c \ll r$ corresponds to $T \gg (a/r)^2 T_c$ ($a \sim 10^{-8}$ cm, $r \sim 10^{-3}$ in magnetic fields $H \sim 10^4$ Oe, $T_c \sim 1°K$--the quantum oscillations are significant only at such temperatures), i.e., $T - T_c \gg 10^{-10}°K$. Violation of this inequality is far beyond the limits of the capabilities of the experiment. At the same time, all the difficulties involved in the construction of the theory of second-order phase transitions pertain to the region $r_c \gtrsim \rho$, if such a region exists.

If $(a/r)^2 \lesssim N^{-1/2}$ (N—number of particles) then the inequality $r_c \ll r$ is satisfied in the entire region of applicability of the thermodynamics, since the measurement of the temperature is meaningful only accurate to the fluctuations: $\tau \gg N^{-1/2}$. (We note that $(a/r)^2 \sim n^{-4/3}$, where n is the density of the Fermi particles.)

Neglecting the fluctuations, the increment caused by the magnetic field in any thermodynamic potential θ depends on the "total" induction. Inasmuch as the inhomogeneous increment near the transition point is small compared with the homogeneous one, θ can be expanded in a series in powers of this increment. Assuming the period λ of the oscillations to be specified[4] (i.e., ensuring the minimum of the thermodynamic potential at a given amplitude of the spatial oscillations A), we obtain an expansion of θ in powers of A, which can be shown (see Sec. 3) to contain only even powers of A and to be analogous to the well-known Landau expansion:

$$\theta = \theta_0 + \alpha A^2 + \beta A^4 + \gamma A^6 + \ldots \qquad (1.1)$$

When $\alpha = 0$ and $\beta > 0$, we obtain the Landau second-order phase transitions (see[7], Secs. 137, 138) with a finite jump of the specific heat c_H (if the transition takes place at a fixed magnetic field H and at a variable temperature) or of the magnetic susceptibility (if H is varied and T is fixed). The relative magnitude of the jump, as follows from simple estimates, is on the order of $(\hbar\Omega/\epsilon_0)^{1/2}$, where $\Omega \propto B$ is the cyclotron frequency and ϵ_0 is the Fermi limiting energy.

The periodic dependence of the quantum oscillations on B^{-1} leads in the homogeneous case to periodicity of the transition in H^{-1} when H is varied at a given T. Here and throughout we assume, for simplicity, that $\mathbf{H} \parallel z$, where z is one of the principal crystallographic axes. At a mean free path $l = \infty$ there is no characteristic length in this direction, and in the main approximation in $r/l \ll 1$ the field is $H = H_z(x, y)$. This means (since curl $\mathbf{H} = 0$) that

$$H = H_z = \text{const}, \qquad M = M_z(x, y), \qquad B = B_z(x, y). \qquad (1.1a)$$

A phase transition is possible, of course, not only from a homogeneous structure to a periodic one, but also when an increment (finite or infinitesimally small) with a new period that differs from the "old" one by a finite amount appears against the background of the inhomogeneous structure with finite amplitude.

The second-order phase transition curve may terminate by crossing the first-order phase transition curve at the critical point of the second-order phase transition (see[7], Sec. 140), where we have in (1.1) $\alpha(T_0, B_0)$

[3] The possibility of appearance of a periodic structure was first indicated in [6]. However, the solution obtained there corresponds to $4\pi\chi > 1$, and is thermodynamically absolutely unstable, and therefore can never be realized physically (see also Sec. 3).

[4] It is finite when $T_c = T$, assuming that the function $\lambda(T)$ is regular, we obtain a temperature increment to λ_c which is linear near T_c. This is proved in Sec. 3.

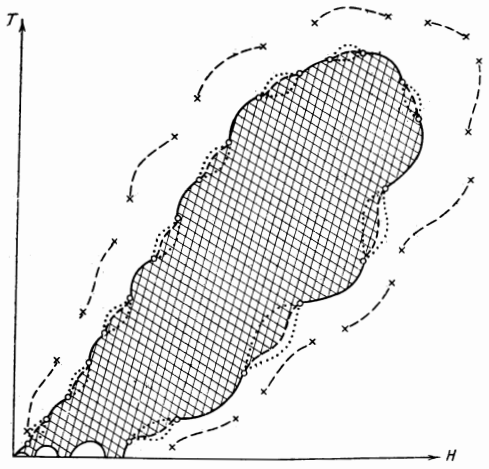

FIG. 1. Diagram of state. Solid line—line of second-order phase transitions, dashed—line of first-order first transitions, dotted—line of absolute instability. ○—critical point of second-order phase transitions, ×—critical point; the shaded area is that of the existence of a spatially-periodic structure.

$= \beta(T_0, B_0) = 0$. With $\alpha = a_1T_1 + b_1H_1$ and $\beta = a_2T_1 + b_2H_1$ (where $T_1 = T - T_0$, $H_1 = H - H_0$, and $|T_1/T| \ll 1$, $|H_1/H| \ll 1$), it is best to test the form (1.1) for a minimum in the coordinate system (α, β), and then transform to the coordinates (T_1, H_1). When $\gamma > 0$ it turns out that the direct continuation of the second-order phase transition curve is the line of absolute instability of the homogeneous phase with A = 0 (see below concerning the investigation of the curve separating the region of the absolute instability); the line of the first-order phase transitions (corresponding to stratification into a homogeneous phase with A = 0 and a periodic phase with $A \neq 0$), and the line of absolute instability of the periodic phase with $A \neq 0$ (see Fig. 1) are tangent to this line at the point (T_0, H_0). The specific heat c_H or respectively the susceptibility χ_T become infinite like $|T_1|^{-1/2}$ or $|H_1|^{-1/2}$.

The phase transitions connected with the diamagnetism take place also when a domain structure appears. In fact, it is clear from Fig. 2 that in an external magnetic field $\mathbf{H}_0$ parallel to the surface of the sample, when H_0 coincides with H in the sample, variation of H_0 is accompanied by a "boiling"—a first-order phase transition from B_c to B_c'. In a field H_0 normal to the surface (Fig. 3) and coinciding with the value of the induction $\overline{B}$ averaged over the sample, an "evaporation" takes place when $B_c \leq H_0 \leq B_c'$, and is accompanied by a jump in the susceptibility[5)] (Landau-type second-order phase transition). In a magnetic field $\mathbf{H}_0$ which is oblique to the surface, it is possible to carry out the most complete investigation by using an additional parameter—the angle of inclination of $\mathbf{H}_0$. Of special interest here is the vicinity of the "critical point," where $4\pi\chi(B_0, T_0) = 1$, i.e., $\partial H/\partial B = 0$, meaning, by virtue of the stability of the state at this point, that $\partial^2 H/\partial B^2 = 0$ and $\partial^3 H/\partial B^3 > 0$.

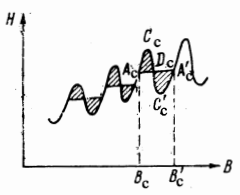

FIG. 2

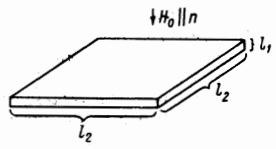

FIG. 3

Putting $B = B_0 + B_1$, $T = T_0 + T_1$, $H = H_0 + H_1$, and taking into account the fact that[6)]

$$\delta\theta = -B\delta H, \tag{1.2}$$

we get

$$\theta = \theta_0 + (a_2T_1 - H_1/4\pi)B_1 + {}^1/_3a_3T_1B_1^2 + {}^1/_4a_4B_1^4. \tag{1.3}$$

This expansion is similar to the Gibbs expansion near the ordinary critical point of the liquid-vapor system (see, for example,[7], Sec. 83), except that the role of the pressure p is played by H, and that of the specific volume V is played by B. (Of course, near the "ordinary" critical point an experimental investigation for a specified connection between p and V, corresponding to an oblique H_0, is extremely difficult.) Therefore all the results of the Gibbs study of the critical point can be directly transferred to our case. Thus, in analogy with formula (84.10) from Sec. 137 of[7], we have

$$c_H \propto (a_3T_1 + 3a_4B_1^2)^{-1}. \tag{1.3a}$$

In particular, on the equilibrium curve, where $B_1 \propto |T_1|^{1/2}$, the specific heat c_H is proportional to T_1^{-1}, and in the case of a "critical" magnetic field ($H_1 = 0$), when it follows from the minimum of θ (i.e., from $\partial\theta/\partial B_1 = 0$) that $B_1 \propto T_1^{1/3}$, formula (1.3a) yields $c_H \propto T_1^{-2/3}$.

In order to distinguish the phase transitions connected with the appearance of the periodic and domain structures, it is necessary to use the essential dependence of the latter on the geometry of the bulky sample, and the independence of the former. Thus, for example, if $\mathbf{H}_0$ rotates in a plane perpendicular to a fourfold axis (the sample dimensions are L_1, L_2, $\sqrt{rL_1} \gg L_2 \gg r$), the character of the phase transitions for $\mathbf{H}_0 \parallel x$ and $\mathbf{H}_0 \parallel y$ will coincide for the periodic structure and differ greatly for the domain structure (first order for $\mathbf{H}_0 \parallel x$ and second order for $\mathbf{H}_0 \parallel y$).

[5)] $\overline{\mathbf{B}} = \mathbf{H}_0$ follows from the continuity of $\int B_n\, dS$, taken over the surface shown in Fig. 3, where $l_2/l_1 \to \infty$ and $l_1 \to \infty$. From $\overline{B} = H_0$ it follows that $H_0 = cB_c + (1 - c)B_c'$, where c is the concentration of phase B_c. The points B_c and B_c' correspond to $4\pi\chi < 1$ and are obtained from the equality of the areas $A_cC_cD_c$ and $D_cC_c'A_c'$, which follows from the minimum of the thermodynamic potential.

[6)] It follows from the foregoing that when $\mathbf{H}_0 \parallel \mathbf{n}$ the independent variable is B, the mean value of which is specified. This means that in the equilibrium state θ is minimal, $\delta\theta = (1/4\pi)H\delta B$, under the condition $\overline{\mathbf{B}} = \mathbf{H}_0$, i.e., that $\theta = \theta + \zeta \int B dr$ has an unconditional minimum, meaning that $(\delta\theta^*/\delta B = 0)$ and $\zeta = -H/4\pi$. Hence $\delta\theta^* = -(1/4\pi)B\delta H$. When $\mathbf{H}_0 \perp \mathbf{n}$, the independent variable is $\mathbf{H} = \mathbf{H}_0$, and we again arrive at formula (1.2). (We note that in the case of a stratification into a liquid and a vapor at a constant value, the independent variable is the average specific volume V; the ambiguity of V(p) makes it possible to vary the pressure p for a specified V, and the minimum is possessed by the thermodynamic potential Φ, the values of which per particle, i.e., the chemical potentials, coincide in both phases.) From (1.2) follows a connection between θ and the "proper" potential (connected only with quantization) of the magnet θ_m:

$$\theta = \theta_m + 2\pi \int M^2\, dr, \qquad M = \frac{B - H}{4\pi} = -\frac{\delta\theta_m}{\delta B}. \tag{1.2a}$$

An important feature of such phase transitions is the macroscopicity of the interaction radius. The finite dimension of the sample leads in this case to a relative width of the transition (both with respect to temperature and with respect to magnetic field), of the order of $(r/D)^2$, where D are the linear dimensions of the sample. Inside this region, all the thermodynamic quantities are rapidly varying but analytic functions.

Observation of phase transitions to an inhomogeneous structure and to such structures in general requires, as is clear from the statement made at the beginning of this section, satisfaction of the condition $4\pi|\chi| \gtrsim 1$, where

$$\chi \sim \left(\frac{v}{c}\right)^2 \left(\frac{\hbar\Omega}{\varepsilon_0}\right)^{-3/2} \exp\left\{-\frac{2\pi^2 T}{\hbar\Omega} - \frac{2\pi^2}{\Omega\tau}\right\} f\left(\frac{\varepsilon_0}{\hbar\Omega}\right); \qquad (1.4)$$

here f(x) is an oscillating function with periods on the order of unity, v is the Fermi velocity of the charges, and τ is their free path time. (This estimate can be obtained also from the formulas of Lifshitz and Kosevich[8] and Bychkov[9].)

Formula (1.4) shows that inhomogeneous structures occur in weak magnetic fields and at low temperatures in pure samples (when T = 0 and $\tau = \infty$ the susceptibility $\chi \to \infty$ as B → 0), when

$$2\pi^2 T \lesssim \hbar\Omega \lesssim \varepsilon_0 (4\pi)^{2/3} (v/c)^{4/3} \qquad \Omega\tau \gtrsim 1, \qquad \Omega \propto H. \qquad (1.5)$$

The interference of different periods leads to a decrease of χ and to a smearing of the temperature of the transition, or in general to the impossibility of an inhomogeneous structure, if χ turns out to be too small. This means that the crystal mosaic structure should be weak and that the magnetic field be highly stable in time and homogeneous in space (the variations of the magnetic field should be small compared with the period ΔH of the oscillations of χ, i.e., with $H(\hbar\Omega/\epsilon_0) \sim H^2$).

The spatial periodic structure will affect many physical properties of the magnet. In the presence of a periodic superstructure, the propagation of electromagnetic and ultrasonic waves in the magnet changes; in particular, when the wave amplitude is sufficiently large, similarities appear, connected with phase transitions that are periodic in time. New types of resonances are produced as a result of new branches of the natural oscillations in the superstructure. A spatial modulation of the specific volume arises (magnetostriction in a periodic field). There appears a periodic electrostatic potential $\varphi(r)$, which ensures conservation of the number of particles in the periodic induction B:

$$e\varphi(r) = -\frac{1}{\nu(\varepsilon_0)} \frac{\delta\theta}{\delta B} \sim \hbar\Omega \left(\frac{\hbar\Omega}{\varepsilon_0}\right)^{1/2} \exp\left\{-\frac{2\pi^2 T}{\hbar\Omega} - \frac{2\pi^2}{\Omega\tau}\right\}$$

(ν—charge density).

Understandably, the periodic structures, diamagnetic domains, and all the effects associated with them are possible also in a ferromagnet, where even in the absence of an external magnetic field we have $B = 4\pi M_0(T)$, where $M_0(T)$ is the spontaneous magnetic moment.

2. DIAGRAM OF STATE

Let us clarify the general form of the diagram of state in a magnetic field, with allowance for the occurrence of periodic and domain structures. At high temperatures $T \gg (2\pi^2)^{-1}\hbar\Omega$, the susceptibility connected with the Landau diamagnetism and the Pauli paramagnetism is small at all temperatures, and an inhomogeneous structure is impossible even in metastable form. (This can be readily shown by using perturbation theory, see also Sec. 3.)

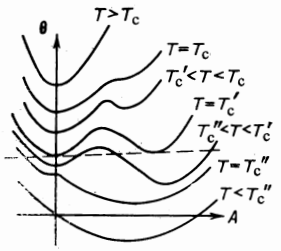

FIG. 4

With decreasing temperature, a local minimum first appears for the inhomogeneous field at a certain temperature. Several different cases are then possible in principle.

The local minimum can appear first at a finite amplitude of the inhomogeneity, so that the smallest value of θ will be ensured as before by the homogeneous induction B. The function $\theta(A)$ will then have at least two minima, between which there must be a relative maximum. Thus, there appear immediately in $\theta(A)$ (besides the minimum in the case of homogeneous B, i.e., when A = 0), also a relative maximum and a minimum, so that when $T = T_c$ of the "creation" of the minimum corresponds to a threefold degenerate solution: $\partial\theta/\partial A = \partial^2\theta/\partial A^2 = 0$, and this determines the temperature T_c and the amplitude at this point (Fig. 4). The appearance of the local minimum means that there can exist a corresponding phase, albeit in unstable form, and its vanishing means absolute instability of such a phase. The curve of the "creation" of the minima on the (T, H) plane thus bounds the region of absolute instability of the given state (in analogy with the $(\partial p/\partial V)_T = 0$ curve in the liquid-vapor stratification).

By starting precisely with this, let us consider the singularity on the curve separating the region of the absolute instability. In the general case it would be necessary to carry out the investigation in similar fashion. For example, in the study of the stratification of a liquid-vapor system it would be necessary to find the thermodynamic potential for a specified inhomogeneous density. The absolute minimum of this functional would determine the equilibrium state, the relative minimum would determine the metastable state, the appearance of a relative minimum would determine the limit of the absolute instability, and the form of the thermodynamic potential near this curve would determine the character of the singularity.

Expanding $\theta(A)$ in powers of $A_1 = A - A_0$ and $T_1 = T - T_c$ (recognizing that $\partial^2\theta/\partial A_1^2 = \partial\theta/\partial A_1 = 0$ and that the expansion is valid because of the same reasons as the expansion (1.1)—see also Sec. 4), we get

$$\theta = \theta_0 + \alpha T_1 A_1 + {}^1/_6 \beta A_1^3. \qquad (2.1)$$

When $\alpha T_1/\beta < 0$, θ has no minimum, when $\alpha T_1/\beta > 0$ there is a relative minimum at $A_1 = (-2\alpha T_1/\beta)^{1/2}$, and $\theta - \theta_0 \sim |T_1|^{3/2}$. This denotes that on approaching the absolute-instability curve at a specified H_0, the specific heat tends to infinity like $|T_1|^{-1/2}$. When this curve is

approached isothermally ($H_0 = H_c + H_1$), the susceptibility goes to infinity like $|H_1|^{-1/2}$.

If then the minimum of θ, becoming deeper, reaches at $T = T_c'$ the same value as in the "preceding" absolute minimum at A = 0, then stratification will take place into a homogeneous phase (A = 0) and an inhomogeneous phase ($A \neq 0$)--a first-order phase transition[7]. When $T < T_c'$, the inhomogeneous state becomes stable, and the homogeneous one metastable at $T_c'' < T < T_c'$ and absolutely unstable at $T < T_c''$ (in the latter case the singularity near $T = T_c''$ is the same as above; of course, we assume that H_0 is specified and not T only for the concreteness).

It may turn out, however, that an instability of the homogeneous state will appear at A = 0 even before the stratification into the homogeneous and inhomogeneous phases takes place (if it does take place at all--the minimum at $A \neq 0$ can start to move upward beyond a certain temperature). Inasmuch as already mentioned and shown in Sec. 3, the expansion of $\theta(A)$ contains only even powers of A, this implies the second-order phase transition considered in Sec. 1 (see Fig. 5); the minima at $\pm A$ correspond to phases with the integral periods and the different "origins" (see Sec. 3). If on the other hand the minima of $\theta(A)$ for different $A \neq 0$ coincide, then stratification takes place into structures having different periods.

Inasmuch as the minima on the $\theta(A)$ curves can move both "upward" and "downward" when the external conditions change (and for a specified T and a change in the magnetic field, their motion is certainly periodic; in general, it follows from (1.5) and from the fact that θ is a superposition of functions that are periodic in B^{-1} that for a specified $T < \hbar\Omega/2\pi^2$ the number of transitions of a given type is of the order of $(\epsilon_0/T)(v/c)^{4/3}/4)$, and different combinations of the aforementioned cases can occur. In particular, a new structure can arise in a first-order transition only at one point (H_{00}, T_0)—see Fig. 6 (the figure shows $\theta(A)$ for two structures: I and II).

The diagram of state shown in Fig. 1 in terms of the variables T and H takes all the foregoing into account; the changeover to the variables T and H_0 is clear from the already indicated connection between H_0 and H. Allowance was made in Fig. 1 for the fact that the phase-transition curve cannot terminate for a periodic structure and can terminate at the critical point for a domain structure; the character of the transitions to the domain structure is the same along the entire curve and depends only on the orientation of H_0 (in Fig. 1 for concreteness, H_0 is perpendicular to the surface of the sample). The form of the diagram of state would be greatly complicated in the next higher approximation in a/r (a--interatomic distance), where the commensurability of a^2 and ehH/c is important (see also[10]).

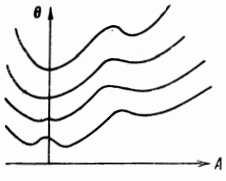

FIG. 5

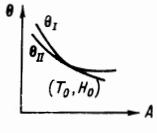

FIG. 6

3. THEORY OF SECOND-ORDER PHASE TRANSITIONS

From (1.2a) and (1.1a) we get the following fundamental equation of the theory of periodic structures:

$$B - 4\pi M\{B\} = H = \text{const}; \quad B = B(\mathbf{R}), \quad \mathbf{R} \equiv (x, y), \tag{3.1}$$

where $M\{B\}$ is a nonlocal functional of B (which relates points separated by a distance on the order of r). The concrete form of $M\{B\}$ is immaterial and will be determined for the magnetic-moment quantum oscillations of interest to us in a separate paper (see also[11]).

Equation (3.1) always has a homogeneous solution B_0:

$$B_0 - 4\pi M(B_0) = H, \tag{3.2}$$

which may turn out to be not unique. Let us ascertain the conditions for the existence of an inhomogeneous solution which is infinitesimally close to the homogeneous one:

$$B = B_0 + B_1. \tag{3.3}$$

We have

$$B_1 = 4\pi\hat{\chi}B_1, \tag{3.4}$$

where $\hat{\chi}$ is a linear integral operator (in accordance with the statement made above). From the homogeneity of the space (with respect to shifts by one-third of the crystal lattice, which in the main approximation is infinitesimally small compared with r), it follows that this operator should be a difference operator, and the invariance of the crystal against inversion ($\mathbf{R} \to -\mathbf{R}$) ensures that this is an even operator, so that (3.4) can be written in the form

$$B_1(r) = \int \chi(\mathbf{R} - \mathbf{R}')B_1(\mathbf{R}')d\mathbf{R}'; \quad \chi(-\mathbf{R}) = \chi(\mathbf{R}), \quad \mathbf{R} \equiv (x, y). \tag{3.5}$$

Putting

$$B_1 = \mathrm{Re}(Ae^{-i\mathbf{kR}}), \tag{3.6}$$

we arrive at the following equation for the period of the spatial oscillations:

$$4\pi\tilde{\chi}(\mathbf{k}_0) = 1, \quad \tilde{\chi}(\mathbf{k}) = \int \chi(\mathbf{r})\cos(\mathbf{kR})d\mathbf{R}. \tag{3.7}$$

It can be shown that the necessary condition for the existence of a real solution of this equation in the stability region, with respect to stratification into diamagnetic domains is $4\pi\chi_0 > 1$ (χ_0 and μ_0 are defined at the beginning of Sec. 1). This excludes, in particular, periodic structures for one zone with one extremal section. The relation between k_x and k_y is determined by the symmetry of the Fermi surface, which for the case $\mu_0 \geq 2$ of interest to us coincides with the symmetry of the crystal lattice (otherwise, for higher symmetry of $\epsilon(\mathbf{p})$, the ratio k_x/k_y would have to be found by taking into account the interaction between the charges and the lattice). Thus, $k_x = k_y$ for a cubic lattice (if H is not parallel it would be necessary to minimize θ to determine k_x/k_y and B_{1x}/B_{1y}).

[7] It is clear therefore that a periodic solution with finite amplitude always arises first in unstable form, which is "inconvenient" compared with the homogeneous one. Therefore the case considered in Sec. 3 of [1] is impossible.

The condition for the solvability of (3.7) is, when $4\pi\chi < 1$, not only the necessary condition but also the sufficient condition for the occurrence of a periodic structure. To prove this, let us expand $\theta\{B\}$ in terms of B_1. Recognizing that (1.2a) should coincide in this case with (3.5), and taking the symmetry of the crystal into consideration, we get

$$\theta = \theta_0 + \frac{1}{8\pi}\int B_1^2(\mathbf{R})\,d\mathbf{R} - \frac{1}{2}\int \chi(\mathbf{R}-\mathbf{R}')B_1(\mathbf{R})B_1(\mathbf{R}')\,d\mathbf{R}\,d\mathbf{R}' + \frac{1}{12\pi}\int f(\mathbf{R}-\mathbf{R}',\mathbf{R}-\mathbf{R}'')B_1(\mathbf{R})B_1(\mathbf{R}')B_1(\mathbf{R}'')\,d\mathbf{R}\,d\mathbf{R}'\,d\mathbf{R}'' + \frac{1}{16\pi}\int g(\mathbf{R}-\mathbf{R}',\mathbf{R}-\mathbf{R}'',\mathbf{R}-\mathbf{R}''')B_1(\mathbf{R})B_1(\mathbf{R}')B_1(\mathbf{R}'')B_1(\mathbf{R}''') \times d\mathbf{R}\,d\mathbf{R}'\,d\mathbf{R}''\,d\mathbf{R}''' + \ldots \quad (3.8)$$

There is no term linear in B_1 in (3.8), because, by virtue of translational symmetry, it should be of the form $K_0(B_0)\int B_1(\mathbf{R})d\mathbf{R}$, and stability against homogeneous perturbations (B_1 = const) denotes $\partial\theta/\partial B_1|_{B_1=0} = K_0(B_0) = 0$ (and $\partial^2\theta/\partial B_1^2|_{B_1=0} = 1 - 4\pi\chi > 0$ by definition).

We rewrite (3.8) in the form

$$\theta = \theta_0 + \frac{\pi}{2}\int (1 - 4\pi\tilde{\chi}(\mathbf{k}))\,|\bar{B}_1(\mathbf{k})|^2\,d\mathbf{k} \quad (3.9)$$

(the tilde denotes here and throughout the Fourier components of the corresponding functions).

If (3.7) has a solution, then $4\pi\tilde{\chi}_{max} \geq 4\pi\tilde{\chi}(\mathbf{k}_0) = 1$, and a small inhomogeneity, according to (3.9), is certainly favored, and the homogeneous state is unstable. As already noted in Sec. 1, the instability of the homogeneous state can be connected either with the appearance of a periodic structure or with the stratification into diamagnetic domains. The latter occurs (see Fig. 2) already when $4\pi\chi < 1$ (i.e., $\partial H/\partial B > 0$); only such values of χ are realized. Therefore, for the existence of a periodic structure it is necessary[8] to have $4\pi\tilde{\chi}_{max} \geq 1$ (the maximum is taken with respect to k) when $4\pi\chi = 4\pi\tilde{\chi}(0) < 1$.

We shall assume that this condition is satisfied. The second-order phase transition point corresponds to the first appearance of the root of (3.7), i.e., $4\pi\tilde{\chi}_{max} = 1$, meaning that a multiple root of (3.7) appears (see Fig. 7). Near this point, the interval of k in which $1 - 4\pi\tilde{\chi} < 0$ is obviously small: $|\Delta k/k_0| \ll 1$. This means (see Sec. 7) that when $|\Delta k| \gtrsim k_0$, where $1 - 4\pi\chi(\mathbf{k}) \gtrsim 1$, the value of B_1 should be small compared with its value in the interval where $1 - 4\pi\tilde{\chi} < 0$, since the term written out in (3.9) is decisive for small values of B_1 (in accordance with the sought assumption), and the state with a small "spread" Δk is certainly not favored. This means that even if account is taken of the next higher terms of the expansion, B_1 near the transition point can be represented in the form

$$B_1(\mathbf{R}) = A(\mathbf{R})e^{i\varkappa\mathbf{R}} + A^*(\mathbf{r})e^{-i\varkappa\mathbf{R}} + C(\mathbf{R}), \quad |C| \ll |A|; \quad (3.10)$$

$$\tilde{\chi}(\varkappa) = \tilde{\chi}_{max}, \quad \nabla\tilde{\chi}(\varkappa) = 0, \quad (3.11)$$

where $A(\mathbf{R})$ is a slowly varying function (over distances large compared with the interaction radius in the kernels of (3.8)—the Larmor radius r), the asterisk denotes the complex conjugate, and $C(\mathbf{R})$ can correspond to any $\mathbf{k}$ and can vary in any manner, but is small compared with $A(\mathbf{R})$.

The form (3.10) allows us to solve by successive approximations[9] the nonlinear equation for $B_1(\mathbf{R})$ which follows from $\delta\theta/\delta B_1 = 0$:

$$B_1(\mathbf{R}) = 4\pi\int \chi(\mathbf{R}-\mathbf{R}')B_1(\mathbf{R}')\,d\mathbf{R}' + \int f(\mathbf{R}-\mathbf{R}',\mathbf{R}-\mathbf{R}'')B_1(\mathbf{R}')B_1(\mathbf{R}'')\,d\mathbf{R}'\,d\mathbf{R}'' + \int g(\mathbf{R}-\mathbf{R}',\mathbf{R}-\mathbf{R}'',\mathbf{R}-\mathbf{R}''')B_1(\mathbf{R}')B_1(\mathbf{R}'')B_1(\mathbf{R}''')\,d\mathbf{R}'\,d\mathbf{R}''\,d\mathbf{R}''' \quad (3.12)$$

Substituting (3.10) in (3.12) we find (in the second approximation in A) the value of C:

$$C = Ee^{2i\varkappa\mathbf{R}} + E^*e^{-2i\varkappa\mathbf{R}} + 2D, \quad (3.13)$$

$$E = \frac{\tilde{f}(\varkappa,-\varkappa)}{1-4\pi\tilde{\chi}(2\varkappa)}A^2, \quad D = \frac{\tilde{f}(\varkappa,-\varkappa)}{1-4\pi\tilde{\chi}(0)}|A|^2. \quad (3.14)$$

In the next approximation, besides the third harmonics, the terms of third order in B_1 gives rise to first harmonics. The corresponding equation (which, of course, coincides with the condition of orthogonality of the "perturbing" inhomogeneous terms in (3.12) to the solution of the homogeneous equation) is

$$-2\pi\sum_{i=1}^{2}\frac{\partial^2 A}{\partial R_i^2}\frac{\partial^2\tilde{\chi}}{\partial\varkappa_i^2} + \tau A - 4\beta A|A|^3 = 0; \quad (3.15)$$

$$\tau = 4\pi\tilde{\chi}(\varkappa) - 1, \quad 2\beta = 3\tilde{g}(\varkappa,\varkappa,-\varkappa) + \frac{2\tilde{f}(\varkappa,2\varkappa)\tilde{f}(\varkappa,\varkappa)}{1-4\pi\tilde{\chi}(2\varkappa)},$$

$$\alpha_i^{-1} = -2\pi\frac{\partial^2\chi}{\partial\varkappa_i^2} > 0. \quad (3.16)$$

Here $\alpha_i > 0$, since χ has a maximum at the point κ; the direction of the axes x and y is chosen such that $\partial^2\tilde{\chi}/\partial\kappa_x\partial\kappa_y = 0$.

We introduce κ_0—the point at which (see Fig. 7)

$$4\pi\tilde{\chi}(\varkappa_0) = 1, \quad \nabla\tilde{\chi}(\varkappa_0) = 0. \quad (3.17)$$

The three equations of (3.17) define, besides κ_0, also the connection between T and H, i.e., the curve of the phase

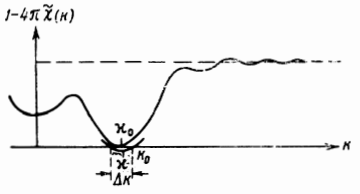

FIG. 7

[8] For a given zone and for a given section, $\chi_\mu(\mathbf{r})$ is of fixed sign, and pick u $\operatorname{sign}\chi_\mu(\mathbf{r}) = \operatorname{sign}\chi_\mu$, $4\pi|\tilde{\chi}_\mu| \leq 4\pi|\tilde{\chi}_\mu(0)| = 4\pi|\chi_\mu|$.

Therefore to realize a periodic solution it is necessary to have several extremal sections and $4\pi\chi_0 > 1$ for $4\pi\chi < 1$.

[9] It is clear that the choice of the zeroth approximation in the form (3.10) already determines completely the course of the successive approximations (of course, with allowance for the condition for the solvability of the inhomogeneous equation in the case when the homogeneous equation has a nontrivial solution). With this, the solution of the equation $\int h(\mathbf{R}-\mathbf{R}')\Phi(\mathbf{R}')d\mathbf{R}' = F(\mathbf{R})\exp(is\cdot\mathbf{R})$ with $F(\mathbf{R})$ a slowly varying function is

$$\Phi(\mathbf{R}) = \exp(is\mathbf{R})H^{-1}\left(i\frac{d}{d\mathbf{R}}\right)F(\mathbf{R}), \quad H(\xi) = \int \exp\{i\mathbf{R}(\xi - s)\}\,h(\mathbf{R})d\mathbf{R}.$$

transitions from the homogeneous to the periodic structure in the (T, H) plane. If the transition is observed at fixed H, then $\tau \sim T - T_0(H)$ ($T_0(H)$ is the transition point), and if T is fixed, then $\tau \sim H - H_0(T)$.

Putting in (3.15) $A = |A|\exp(i\psi)$, we find that if $\psi \neq 0$, then $\psi \sim |A|^{-2}$ and is a function that oscillates rapidly in R (since |A| is small by definition), which, as shown above, is incorrect. Therefore $\psi = 0$ and A is real. Substituting (3.10) with real A in (3.9) we get

$$\theta - \theta_0 = \int \left\{ -U(A) + \sum_{i=1}^{2} \frac{1}{2a_i} \left(\frac{\partial A}{\partial R_i} \right)^2 \right\} d\mathbf{R} \equiv \int \theta_1 d\mathbf{R},$$
$$U(A) = {}^1/_4 \tau A^2 - {}^1/_2 \beta A^4. \qquad (3.18)$$

As mentioned in Sec. 1, there are no terms of first and third order in A in (3.18).

Since, as follows from the foregoing general considerations, a periodic structure with period $2\pi\kappa_i^{-1} \sim r$ occurs at the transition point, the form (3.18) of the expansion of θ in terms of the slowly varying increment A(R) (compared with r) can be obtained directly. To this end it is necessary to take into consideration the following: 1) from the requirement that θ have a minimum (already averaged over distances of the order of r) with respect to A at the transition point it follows that $\delta\theta/\delta A = 0$; 2) $\delta H/\delta A \propto \tau$ (since $4\pi\tilde{\chi}(\kappa_0) = 1$), and this yields $\delta^2\theta/\delta A^2 \propto \tau$; 3) the presence of the minimum of $\theta(A)$ at $T = T_0$ calls for $\delta^3\theta/\delta A^3 = 0$ and $\delta^4\theta/\delta A^4 = \beta > 0$; 4) the expansion in terms of the small ∇A can contain, owing to the slow variation of A(R) can contain only even powers of ∇A (in view of the invariance with respect to the replacement of R by $-R$).

Let us explain the meaning of the requirement $\beta > 0$ in our case. If $\beta < 0$, then it follows from (3.18) that the point $\tau = 0$ is not at all singular—there already exists in it a periodic structure with a finite amplitude, the transition occurred earlier, and furthermore at finite A (since it is the transition $A \to 0$ which we are investigating), i.e., we have a first-order phase transition.

Since $\theta_i > 0$ (see (3.16)), it follows that $\theta_1 \geq -U(A) \geq -U_{max}$, and the equality $\theta_1 = -U_{max}$ is attained for homogeneous A. With this, the term with the derivative in (3.18) vanishes, and θ_1 takes on a form characteristic of Landau-type second-order phase transitions[10] (see (1.1)) and leads to a transition from $A = 0$ when $\tau < 0$ to $\pm A_0$, $A_0 = (1/2)\sqrt{\tau/\beta}$ when $t > 0$. States with $\pm A_0$ differ only in a phase shift. This difference can be appreciable in a finite sample (where such states are analogous to domains). In determining κ and κ_0 it is easy to find the dependence of the spatial periods $2\pi\kappa_i^{-1}$ on τ: $\kappa - \kappa_0 \propto \tau$. Thus, the period of the oscillations varies linearly in τ close to the transition points, and their amplitude is proportional to $\sqrt{\tau}$.

All the arguments presented above are particularly clear in the one-dimensional case (A = A(y)), when the functional (3.18) can be formally interpreted as the action for a one-dimensional motion of a particle of mass α in a time y along the coordinate A (see also[11]) with a forbidden departure of the particle to infinity (since B should be finite). The transition between the states with $\pm A$ has in this case a "domain" character and occurs (when $\tau > 0$) in accordance with the law $A = A_0 \tanh\{(1/2)\sqrt{\tau\alpha}(y - y_0)\}$.

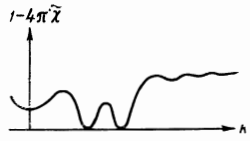

FIG. 8

From the definition of (3.7) it is clear that as $k \to \infty$ the function $\tilde{\chi}(k)$ oscillates and tends to zero. This means that when the temperature (or magnetic field) is changed, there should appear new roots of the equation (3.11) and accordingly new phase transitions should take place. If any extremum of $\tilde{\chi}(k)$ with $k > 0$ turns out to be degenerate (Fig. 8), stratification into phases with different periods is possible.

4. THE CRITICAL POINT AND THE CURVE OF ABSOLUTE INSTABILITY

A singular point is a point of obligatory degeneracy of κ, i.e., $\kappa_0 = 0$ (we recall that $\tilde{\chi}(-\mathbf{k}) = \tilde{\chi}(\mathbf{k})$ and therefore the point $k = 0$ must correspond to an extremum). According to (3.7) we have here $4\pi\chi = 1$. In this case we can use the theory developed in the preceding section, but it is simpler to note immediately that when $\kappa_0 = 0$ the entire quantity B varies slowly, and therefore $\theta_1\{B\}$ can be expanded in powers of ∇B. In the fundamental approximation, θ_1 coincides with the "local" homogeneous density $\theta_1^0(B)$, the form of which was written out earlier (formula (1.3)). In the next higher approximation,

$$\theta_1\{B\} = \theta_1^0(B) + \sum_{i=1}^{2} \frac{1}{2a_i} \left(\frac{\partial B}{\partial R_i} \right)^2.$$

Reasoning just as in Sec. 2, we can verify that the critical point pertains to stratification into higher magnetic domains. Outside the domain wall, the derivatives in $\theta_1\{B_1\}$ should be discarded, and the argumentation that follows is the same as in Sec. 1. (The shape of the domain wall for the one-dimensional case was obtained in[5]).

Let us turn to explain the character of the singularity of the thermodynamic quantities on the absolute-instability curve (where in the homogeneous case $\partial H/\partial B = 0$). Assume that when H and $T = T_0$ are specified, the equation for B, which we shall write symbolically in the form

$$\hat{L}\{T, H; B\} = 0, \qquad (4.1)$$

has a particular solution $B_0(R)$ such that

$$\hat{L}(T_0, H; B_0(\mathbf{R})\} = 0. \qquad (4.2)$$

Let us ascertain the character and the stability of the solution at $T = T_0 + T_1$. Putting $B = B_0 + \psi$, we get

$$\hat{L}_1\psi + T_1\hat{L}_2\{B_0(\mathbf{R})\} + {}^1/_2\hat{L}_3\psi^2 + \ldots = 0, \qquad (4.3)$$

where

$$\hat{L}_1 = \frac{\delta\hat{L}}{\delta B_0}, \quad \hat{L}_2\{B_0(\mathbf{R})\} = \frac{\partial}{\partial T_0}\hat{L}\{T_0, H; B_0(\mathbf{R})\}, \quad \hat{L}_3 = \frac{\delta^2 L}{\delta B_0^2}. \qquad (4.4)$$

[10] It is clear from the foregoing investigation that the third order phase transition referred to in [1] cannot precede a second-order phase transition, in which supercooling or superheating is impossible, and therefore is not realized.

We vary T for a specified H is only for concreteness, and the analysis is perfectly similar for specified T and variable H.

The solution and investigation of (4.3) are performed in exactly the same manner as before for (3.12), and its character is determined by the presence or absence of a solution of the homogeneous equation $\hat{L}_1\psi = 0$; the point of T where such a solution first appears gives the $T_0 = T_0(H)$ curve. The only difference is that the terms of the thermodynamic potential that are cubic in the amplitude can either vanish (by virtue of the symmetry or in points that are isolated in H), or remain (since, unlike (3.12), the kernels in (4.3) need not be difference kernels, since the inhomogeneity of $B_0(R)$ violates, in general, the translational symmetry in the system). In the latter case, the expansion of the thermodynamic potential in powers of slowly varying A takes on the form (2.1), and the subsequent reasoning is the same as in Sec. 2.

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