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Cholesteric liquid crystal in a magnetic field near the phase transition into a smectic-A

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The dependence of the period of the cholesteric helix on the magnetic field perpendicular to the axis of the field is calculated above the point of the phase transition into the smectic-A. The dependence of the critical field H_c on the temperature is obtained.

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The behavior of a cholesteric liquid crystal in a magnetic field perpendicular to the axis of the cholesteric helix was investigated both theoretically^[1] and experimentally.^[3] With increasing magnetic field, period of the cholesteric structure increases and above the critical field H_c the crystal has nematic ordering. In weak magnetic fields, the period increases like the fourth power of the field and diverges logarithmically near the critical field. We examine this problem from the point of view of the phase transition of a cholesteric liquid crystal into a smectic one. In the phase transition into

the smectic-A phase, the order parameter is the Fourier component ψ of the crystal density, with wave vector $\mathbf{q}_0 = 2\pi\mathbf{n}_0/d$, where d is the distance between planes and \mathbf{n}_0 is the average director and is perpendicular to the equidistantly disposed layers. To take into account the quadratic fluctuations of the order we use the de Gennes Hamiltonian.^[4]

$$H = \int d^3r \left\{ a\rho^2 + \frac{1}{2m_{\parallel}} |(\mathbf{n}\nabla - iq_0)\rho|^2 + \frac{1}{2m_{\perp}} [n\nabla\rho]^2 \right\}, \quad (1)$$

$$\mathbf{n} = (\cos \varphi(z), \sin \varphi(z), 0),$$

where ρ is the deviation of the crystal density from the mean value ρ_0 .

The director \mathbf{n} changes little over the correlation length ξ_1 along the z direction of the axis of the cholesteric helix ($\varphi' \xi_1 \ll 1$). To calculate the free energy in the region $\xi_1 \ll \Delta z \ll (\varphi')^{-1}$ we present ρ in the form

$$\rho = \text{Re}(\psi \exp(iq_0 n_0 r)), \quad (2)$$

where \mathbf{n}_0 is the average direction of director \mathbf{n} in the region Δz . The Hamiltonian (1) then takes the form

$$H = \int d^3r \left\{ a |\psi|^2 + \frac{1}{2m_\perp} |\nabla_\perp \psi|^2 + \frac{1}{2m_\parallel} |\nabla_\parallel \psi|^2 + \frac{1}{2m_\perp} |(\nabla_\perp - iq_0 \delta n_\perp) \psi|^2 + \frac{b}{2} |\psi|^4 \right\}, \quad (3)$$

where

$$\delta \mathbf{n} = (0, \varphi' z, 0), \quad \delta n_\parallel = n - n_0,$$

and \mathbf{n} is directed along the x axis.

The quadratic fluctuations of the order parameter ψ are taken into account in analogy with [4]. For example, for the moment $M = \delta F / \delta \varphi'$ we obtain the expression

$$M(z) = (\varphi')^{1/2} (2q_0)^{1/2} \left(\frac{m_\parallel}{m_\perp} \right)^{1/2} \frac{T}{4\pi} f(\gamma(z)), \quad (4)$$

where now $\gamma(z) = a m_\perp / q_0 \varphi'$.

Formulas (2)–(4) are valid under the assumption $\varphi' \xi_1 \ll 1$ and $\varphi'' \xi_1^2 \ll \varphi' \xi_1$. Indeed, $\xi_1^{-2} \approx 2m_\perp a + q_0 \varphi'$ and $(\varphi' \xi_1)^2 = (2\pi)^{-1} \varphi' d / (2\gamma + 1) \ll 1$. In our problem $\varphi'' \ll \pi^2 / \varphi' p$ where p is the period of the cholesteric structure in the absence of a magnetic field. Therefore the second condition is also satisfied.

The value of the parameter γ is significant for the pre-transition behavior of cholesteric liquid crystals. In contrast to the standard cholesteric liquid crystals, where $\gamma \sim 1$ even in the pre-transition region, [5] the solution of a cholesteric in a nematic, used in [3], has a larger period p , and this leads to a larger value of γ :

$$\gamma = \frac{\xi_{10}^{-2} d p (T - T_c)}{(2\pi)^2 T_c} \approx 1.5 \cdot 10^4 \frac{(T - T_c)}{T_c}$$

where $\xi_{10} = 3 \text{ \AA}$, $d = 30 \text{ \AA}$, and $p = 2 \times 10^5 \text{ \AA}$.

The free energy calculation in analogy with [5] is valid if the Ginzburg parameter is small:

$$\text{Gi}(\gamma) = \langle |\psi|^2 \rangle / |\psi_0|^2.$$

In the limit of large γ , the Ginzburg parameter is the same for a cholesteric and a nematic:

$$\text{Gi}(\gamma \gg 1) = \frac{T b m_\perp m_\parallel^2}{2\pi a^2}, \quad \frac{\delta p}{p_0} = \kappa^{-2} \text{Gi} \frac{\sqrt{2}}{24}, \quad (5)$$

where $\kappa^{-2} = q_0^2 / b K_{22} m_\perp^2$. At $|\nu| \ll \frac{1}{2}$ we have

$$\text{Gi} \left(|\gamma| \ll \frac{1}{2} \right) = \frac{(1 - \sqrt{2}) \xi^{1/2} T b m_\perp m_\parallel^2 \gamma^2}{\pi \sqrt{2} a^2}.$$

On the other hand, at the point of the phase transition in-

to a smectic-A, the energies of the high-temperature and low-temperature stages are equal:

$$1/2 K_{22} (\alpha_c - \alpha_0)^2 + \delta F(\alpha_c) = -a_c^2 / 2b + 1/2 K_{22} \alpha_0^2,$$

where $\delta F(\alpha_c)$ is the contribution from the fluctuations of the order parameter ψ .

In the region $|\gamma_c| \ll \frac{1}{4}$, $|\gamma| \ll \frac{1}{4}$ we represent δF in the form

$$\delta F(\alpha) = \delta K_{22} (\gamma \alpha_0 a^2)^{1/2} = K (\alpha_c \alpha^2)^{1/2},$$

where $\delta p_c / p_0 = 3K / 4K_{22}$. We then obtain for κ an estimate in the form

$$\kappa^{-2} = (2\delta p_c / 3p_0 + 1) \gamma_c^{-2}. \quad (6)$$

We note that in crystals with large period p , such as investigated in [3], the pre-transition growth of p at large γ is defined by

$$\frac{\delta p}{p_0} = \left(\frac{2p m_\perp}{d^2 m_\perp} \right)^{1/2} f(\gamma) \frac{T}{K_{22}} \approx \frac{1}{\gamma^{1/2}} \quad (7)$$

$$\delta p_c / p_0 \approx 2, \quad |\gamma_c| \ll 1/2.$$

It follows therefore from (5)–(7) that

$$\text{Gi} = \frac{72 p_0 \gamma_c^2 \gamma^{-1/2}}{\sqrt{2} (2\delta p_c + 3p_0)} \ll 1$$

at large γ and $|\gamma_c| \ll \frac{1}{4}$. At $|\gamma_c| \ll \frac{1}{4}$ we have

$$\text{Gi} \left(|\gamma| \ll \frac{1}{2} \right) = \frac{3(1 - \sqrt{2}) \xi^{1/2} \delta p_c \gamma_c^2}{f(\gamma_c) (2\delta p_c + 3p_0)} \ll 1.$$

The fluctuations of the order parameter ψ change the free energy of the cholesteric in a magnetic field. This energy was, far from the phase transition into the smectic, of the form

$$F_0 = \frac{1}{2} \int d^3r \{ K_{22} (\varphi' - \alpha_0)^2 - \chi H^2 \sin^2 \varphi \},$$

where $\mathbf{H} = (0, H, 0)$. The Lagrange equation for $F_0 + \delta F$ is written in the form

$$K_{22} (\varphi')^2 + 2\delta M(\varphi') \varphi' - 2\delta F(\varphi') + \chi H^2 \sin^2 \varphi = \chi H^2 k^{-2}. \quad (8)$$

Assuming the nonlinear term of (8) to be small, we linearize (8) and determine its solution in the form

$$\varphi'(\varphi, k) = \varphi_0'(\varphi, k) + \delta \varphi'(\varphi, k), \quad (9)$$

where

$$K_{22} \varphi_0' \delta \varphi' = \delta F(\varphi_0') - \delta M(\varphi_0') \varphi_0',$$

$$\xi^2 (\varphi_0')^2 + \sin^2 \varphi = 1/k^2, \quad \xi^2 = K_{22} / \chi H^2.$$

Then the free-energy density g takes the form

$$\frac{2g}{K_{22} \alpha_0^2} = 1 - \frac{1}{k^2 \xi^2 \alpha_0^2} - \frac{4\pi}{\alpha_0 p} \left(1 - \frac{2E(k)}{\pi \xi \alpha_0 k} \right) + \frac{8}{K_{22} p \alpha_0^2} \int_0^{\pi/2} d\varphi \frac{\delta F(\varphi_0')}{\varphi_0'}, \quad (10)$$

where

$$p = 4k\xi K(k) + \delta p, \quad \delta p = -4 \int_0^{\pi/2} d\varphi \delta\varphi' / (\varphi_0')^2. \quad (11)$$

We shall next deal with the region $p^{-1}\partial p/\partial k \gg 1$, i.e., as $k \rightarrow 1$. The value $k=1$ corresponds to a transition of a cholesteric into a nematic at any $a > 0$. We seek the solution in the form $k = k_0 + \delta k$, minimizing g with respect to the parameter k :

$$2\tilde{\xi} E(k_0) = \pi \tilde{\xi}^2 \alpha_0 k_0, \quad (12)$$

$$\frac{p}{p_0} = \left(\frac{2}{\pi}\right)^2 \left[\left(\frac{\tilde{\xi}}{\xi}\right)^2 K(k_0) E(k_0) + \frac{E^2(k_0) \delta k}{(1-k_0^2)k_0} \right],$$

where

$$\delta k = \frac{2\xi^2 k_0^3}{K_{22} p} \int_0^{\pi/2} d\varphi \frac{2\delta F(\varphi_0') - \delta K_{22} (\varphi_0')^2}{\varphi_0'}, \quad (13)$$

$$\tilde{\xi}^2 = (K_{22} + \delta K_{22}) / \gamma H^2.$$

The integrand in (13) is not singular as $k \rightarrow 1$, inasmuch as at small φ_0' it begins with $(\varphi_0')^2$. We must therefore set $k=1$ under the integral sign in (13). In expression (11) for δp , the singular contribution as $k \rightarrow 1$ is made by the principal term of the expansion $\delta F = \delta K_{22} (\varphi_0')^2/2$, which is included in the first term of formula (12). Formula (12) is valid so long as the second term is small. Formulas (9)–(12) are valid if the linearization condition is satisfied, i.e., if it is possible to neglect in the free energy terms of order

$$\Delta F \sim F_0 \left(\frac{\delta p}{p_0}\right)^2 \ll F_0, \quad \left(\frac{\delta p}{p_0}\right)^2 \ll \frac{1}{10}, \quad \gamma \geq 10. \quad (14)$$

Thus, linearization of (9) is possible at $\gamma \geq 10$. At $\gamma < 1$, first, the possibility of linearization of (9) is lost by virtue of (14) and, second, the Ginzburg parameter for the regions of a cholesteric with small φ' (as $\varphi \rightarrow \pi/2$), where the crystal hardly differs from a nematic liquid crystal, increases like $\gamma^{-1/2}$, and we cannot confine ourselves to the quadratic fluctuations of the order parameter ψ . The region of φ close to $\pi/2$ makes the principal contribution to the period p , but plays no importance in the determination of the H_c shift due to the small contribution to the integral:

$$\frac{\delta H_c}{H_c} = - \left(\frac{2}{\pi}\right)^2 \frac{1}{K_{22} \alpha_0^2} \int_0^{\pi/2} d\varphi \frac{\delta F(\pi \alpha_0 \cos \varphi/2)}{\cos \varphi}. \quad (15)$$

For the crystals used in [5], formula (15) is therefore valid at $\gamma \geq 10$, and its validity is limited only by the linearization condition (14). For ordinary liquid crystals with $K_{22} \approx 10^{-6}$ erg-cm and $p \approx 3 \times 10^3$ Å we have $\delta p/p_0 \ll 1$ in the region $\gamma > 0$, and formula (15) is valid for any $a > 0$. There is no complete untwisting of the cholesteric helix in the magnetic field at $a < 0$, since the nematic structure is unstable to a transition into the smectic-A at $a < 0$. A decrease of φ' leads to an increase of the absolute value of $\gamma(z)$, and instability sets in at the point $\gamma(z) = -\frac{1}{2}$. [5]

At $\gamma \lesssim 1$, regions with small φ_0' make a small contribution to (15). Therefore they calculate the shift of H_c we can use for δF expressions in the form

$$\delta F(\varphi') = \frac{\delta K_{22} (\varphi')^2}{(4 + \gamma^{-1}(z))^{1/2}}.$$

Then

$$\frac{\delta H_c}{H_c} = - \frac{2\delta K_{22} \gamma_0^{1/2}}{\pi^{1/2} K_{22}} \{2E(\eta, r^{-1}) - F(\eta, r^{-1})\}, \quad \frac{8\gamma_0}{\pi} < 1,$$

and

$$\frac{\delta H_c(a=0)}{H_c} \approx \frac{\delta K_{22} \gamma_0^{1/2}}{K_{22}}, \quad \eta = \arcsin\left(\frac{\pi}{\pi + 8\gamma_0}\right)^{1/2}, \quad r = \left(\frac{2\pi}{\pi + 8\gamma_0}\right)^{1/2}.$$

If we take the first terms of the expansion of $F(\varphi')$ in small $\gamma^{-1}(z)$, then the problem can be solved exactly:

$$\delta F = \delta K_{22} (\varphi')^2/2 - \delta K_{22} (\varphi')^2/2\gamma(z), \quad (16)$$

$$\frac{p}{p_0} = \left(\frac{2}{\pi}\right)^2 \left[\left(\frac{\tilde{\xi}}{\xi}\right)^2 K(k_0) E(k_0) + \frac{\delta \bar{p} (2 - k_0^2) E^2(k_0)}{4p(1 - k_0^2)} \right] + \frac{\delta \bar{p}}{p_0}, \quad (17)$$

where

$$\frac{\delta \bar{p}}{p_0} = - \frac{\delta K_{22}}{K_{22} \gamma_0}, \quad \gamma_0 = \frac{m_{\perp} a}{q_0 \alpha_0}. \quad (18)$$

The first term in the expansion (16) of δF leads to simple temperature renormalizations of K_{22} and of the period p_0 . The functional dependence of p on H remains unchanged. The appearance of the next term in the expansion of δF leads to a change in the $p(H)$ dependence. The shift of H_c is equal to

$$\frac{\delta H_c}{H_c} = - \frac{\delta K_{22}}{2K_{22}} \left(1 - \frac{\pi^2}{8\gamma_0}\right),$$

where the first term is the shift of H_c due to the condition (18), and the second is the contribution of the second term of (16). The experiment far from the point of the phase transition into the smectic-A was performed on mixtures of nematic and cholesteric crystals having large periods $p = 2 \times 10^5$ Å with small critical fields $H_c \approx 10^4$ G. For such crystals we have $K_{22} \approx 3 \times 10^{-7}$ erg-cm. Using values typical of cholesterics, $d = 30$ Å and $m_{\parallel}/m_{\perp} = 0.1$, we obtain

$$\frac{\delta K_{22}}{K_{22}} = \frac{\delta p}{p_0} \approx \frac{1}{\gamma_0^{1/2}}$$

at large γ_0 .

In the pre-transition region $\gamma_0 \sim 10$ the second term in (17) becomes noticeable. For example, the change of the period $p - p_0$, due to the second term assumes near H_c the value

$$\frac{p - p_0}{p_0} = - \frac{y^2}{64\gamma_0^{1/2} \ln y}, \quad y = \frac{4H_c^{1/2}}{[2(H_c - H)]^{1/2}}$$

and $p - p_0 \approx -p_0/6$ for $\ln y = 4$ and $\gamma_0 = 10$.

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He³-He⁴ solutions and other low-density Fermi liquids in a magnetic field

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A theory is proposed for a Fermi liquid of low density, such as a solution of He³ in superfluid He⁴, in an arbitrary magnetic field. The characteristics of the solution are determined by an expansion in the concentration $x^{1/3}$ and are specified, in a field that is not too strong, by a single parameter, the s -scattering length. At almost total polarization of the spin system, the main contribution is already made by p -scattering. Values are obtained for the thermodynamic and hydrodynamic quantities and for the propagation velocity of the low-frequency oscillations (transverse spin waves, high-frequency first sound, and coupled spin-sound modes that exist in weak magnetic fields). The most noticeable are the magnetokinetic effects connected with the appreciable growth of the fermion mean free path and of the kinetic coefficients.

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1. INTRODUCTION

The connection between the Fermi-liquid function of a system in an external magnetic field H and its value in the absence of a field has been established only for weak magnetic fields $\beta H \ll T_F$ (β is the magnetic moment of the fermions and T_F is the degeneracy temperature), when the corrections to the f -function are small to the extent that the field is weak. A consistent calculation of the Landau f -function^[1] in the absence of a magnetic field can be carried out in the case of a low-density Fermi system with the aid of an expansion in $x^{1/2}$ ($x \ll 1$ is the concentration of the fermions). All the properties of such systems are then specified by a single interaction parameter—the s -scattering length a of the Fermi particles.^[2-7]

In this paper we propose a theory of an uncharged low-density Fermi liquid in an arbitrary magnetic field with account taken also of the possible presence of a superfluid background. The analysis is based on the fact that for low-density Fermi systems the expansion in the interaction coincides formally with the expansion of all the quantities in powers of $x^{1/3}$. The interaction of the bare particles in the nonrelativistic approximation does not depend on the spin, and the scattering amplitude does not depend on the magnetic field. This makes it possible to relate, in first approximation in the interaction, the f -function of the isotropic Fermi liquid with the amplitude of particle scattering in the absence of a field.

In an isotropic Fermi liquid of spin-1/2 particles, the Fermi surfaces of quasiparticles with differently oriented spins constitute two Fermi spheres whose radii are determined by the degree of polarization of the spin system, i.e., by the value of the magnetic field. In the course of the interaction, the particles with the two

polarizations remain on their Fermi surfaces. If the particle interaction energy decreases rapidly enough with increasing distance between them, then the scattering amplitude of the slow particles, and consequently the low-density Fermi-liquid function, are determined by the s -scattering.

Since the fermions are identical, only particles with oppositely directed spins interact in the s -scattering. In strong magnetic fields, when practically all the spins have the same orientation, the Fermi-liquid interaction is already determined by the p -scattering.^[8] This weakens considerably the interaction in strong fields and alters substantially the concentration dependences of all the thermodynamic quantities. For this reason, the values of the kinetic coefficients, such as viscosity, thermal conductivity, and others, increase strongly with increasing magnetic field (with increasing degree of polarization of the fermions).^[9]

The most typical example of a low-density Fermi liquid is the degenerate solution of He³ in superfluid He⁴, in which an important role is played also by the interaction of the fermions with the superfluid background that moves with velocities v_s .^[9] The calculations that follow have therefore been performed for degenerate He³-HeII solutions.

In the next section of the paper we derive general relations that connect the density matrix, the energy spectrum, and the f -function of a Fermi system in the magnetic field in the presence also of a superfluid background. We consider next the thermodynamic properties of a partially as well as fully polarized Fermi liquid (Sec. 3), high-frequency spin and acoustic oscillations (Sec. 4), and magnetokinetic phenomena (Sec. 5). In the Conclusion we present numerical estimates, and discuss the limits of applicability of the theory and the ex-