Magnetic properties of layered superconductors with weak interaction between the layers

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(Submitted December 20, 1972)
Zh. Eksp. Teor. Fiz. **64**, 2241-2247 (June 1973)

The magnetic properties of layered superconductors with Josephson interaction between the layers is considered. Differential-difference equations for the Ginzburg-Landau parameter in a magnetic field are obtained near the critical temperature. The vortex state in fields parallel to the layers is investigated on basis of the equations. The superconducting state is not destroyed in the center of the vortex filament at temperatures not too close to T_c, but at the filament periphery the conditions are very similar to those in a vortex filament of ordinary strongly-anisotropic type-II superconductors. $H_{c1}(\parallel)$ is calculated and the structure of the vortex scale is considered. It is shown that at temperatures not too close to T_c the magnitude of H_{c2} is determined by the paramagnetic effect and orbital motion of electrons within the layers in a field $H_\perp$. Thus in pure layered superconductors in parallel fields and at low temperatures an inhomogeneous state should exist and the field of transition to this state from the normal state at zero temperature equals $\sqrt{2}H_p(0)$.

It was proposed in[1-3] that a Josephson interaction of the layers can be realized in layered superconductors. We shall assume that the single-electron energy ϵ in the normal state of such compounds can be described by the relation

$$\varepsilon(\mathbf{p}, q) = \mathbf{p}^2/2m + 2b\cos q,\ 0 \leqslant q \leqslant 2\pi, \tag{1}$$

where p is the quasimomentum along the layers, m is the effective mass, q is the quasimomentum for the motion of the electrons between the layers along the z axis (in (1) and below we describe the motion of the electron between layers by using the Wannier representation with the function $w_n(z)$, where n is the number of the layer). The Josephson interaction of the layers or, in other words, the weak superconductivity between the layers, is realized if

$$b \ll T_c \sim \Delta(0). \tag{2}$$

It is possible that condition (2) is realized in layered compounds of dichalcogenides of the type TaS_2 or in their intercalated compounds[4]. If this is not so, then it seems that if the molecules are suitably intercalated then condition (2) can be realized.

In layered superconductors with weak conductivity between the layers (we shall henceforth take layered superconductors to mean only superconductors of this type), at $b \ll \Delta(T)$, i.e., at temperatures

$$\tau = (T_c - T)/T_c \gg b^2/T_c^2, \tag{3}$$

there will be observed effects that are qualitatively similar to those taking place in Josephson junctions. Thus, at a constant potential difference V applied perpendicular to the layer, one can observe an alternating Josephson current at a frequency $\omega = 2eV/\hbar\mathcal{N}$, where $\mathcal{N}$ is the number of layers between which the voltage is applied. In layered superconductors one can also observe natural oscillations corresponding to Josephson plasma oscillation[2]. The features of the magnetic properties of layered superconductors are determined by the fact that in magnetic fields $H_\parallel$ parallel to the layers, when condition (3) is satisfied, the superconductivity cannot be destroyed by the orbital motion of the electrons (owing to the smallness of the Josephson currents between the layers). Therefore the usual picture of the vortical state of type-II superconductors does not apply to layered superconductors in the temperature region (3) (the presence of a normal state at the center of the vortex is not favored energywise). In strong fields $H_\parallel$, for the same reason, the destruction of the superconductivity can occur only as a result of the paramagnetic effect.

GINZBURG-LANDAU DIFFERENTIAL-DIFFERENCE EQUATIONS FOR LAYERED SUPERCONDUCTORS

In the derivation of the equations for the Ginzburg-Landau order parameter in the region $\tau \ll 1$, we employ the customary procedure, using the integral equation

$$\Delta(\mathbf{r}) = K\left(-i\nabla - \frac{2e}{\hbar c}\mathbf{A}\right)\Delta(\mathbf{r}) \tag{4}$$

(A is the vector potential) and add to it terms of third order in Δ (see Chap. 5 of[5]). The transition from the representation r = (x, y, z) to the Wannier representation (x, y, n) with respect to the coordinate z, and the subsequent transition to the momentum representation (p, q), yields for the kernel K the following expressions:

$$K(p, q) = K(p, 0) - K(0, 0) + K(0, q) - K(0, 0) + K_t(0, 0), \tag{5}$$

where $K_t(0, 0)$ is the kernel K with cutoff with respect to the frequencies, and

$$K(p,0) = 2\lambda N(0)\pi T \sum_\omega \operatorname{Im} \int_{-\infty}^{+\infty} d\Omega \frac{g_\parallel(p,\Omega)}{2i|\omega| + \Omega},$$

$$K(0,q) = 2\lambda N(0)\pi T \sum_\omega \operatorname{Im} \int_{-\infty}^{+\infty} d\Omega \frac{g_\perp(q,\Omega)}{2i|\omega| + \Omega}, \tag{6}$$

$$a^{-1} = \int_{-\infty}^{+\infty} |w_n(z)|^4 dz, \quad N(0) = m/2\pi a$$

λ is the interaction constant and N(0) is the density of states. For "clean" (c) and "dirty" (d) superconductors (the respective mean free path $l_\parallel$ in the layer is $l_\parallel \gg \xi_0$ and $l_\parallel \ll \xi_0$) the functions g are determined by the expressions

$$g_\parallel^{(c)}(p,\Omega) = \begin{cases} [\pi^2\hbar^2(p^2v_F^2 - \Omega^2)]^{-1/2}, & |\Omega| \leqslant pv_F \\ 0, & |\Omega| > pv_F \end{cases}, \tag{7}$$

$$g_\parallel^{(d)}(p,\Omega) = \frac{1}{\pi\hbar}\frac{D_\parallel p^2}{D_\parallel^2 p^4 + \Omega^2}, \tag{8}$$

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where v_F is the Fermi velocity of the electron and $D_{\|}$ is the diffusion coefficient of the electron inside the layer in the dirty superconductor.

In the case when the electron motion between the layers is of the band type (b), i.e., $l_\perp$ is large in comparison with the distance between the layers d, and the description (1) is valid, the expression for $g_{\|}(q, \Omega)$ is obtained from (7) by replacing pv_F by $4b\sin(q/2)$.

For the hopping mechanism of electron motion between layers (h), the expression for $g_\perp^{(h)}(q, \Omega)$ is obtained by replacing $D_{\|}p^2$ with $2D_\perp(1 - \cos q)/d^2$, where $D_\perp$ is the coefficient of diffusion of the electron between layers. In this case, the weak-superconductivity condition between layers is given by

$$\hbar D_\perp / d^2 \ll T_c \sim \Delta(0), \tag{9}$$

since $D_\perp/d^2$ is the reciprocal time of the hopping of the electron between layers.

From (4)–(8), accurate to terms of second order in b/T_c inclusive, we obtain for the Ginzburg-Landau parameter $\psi_n(x, y) = |\psi_n(x, y)|\exp[i\varphi_n(x, y)]$, and for the current density the differential-difference equations

$$\begin{gathered}
\left[\frac{\hbar^2}{2m}\left(-i\nabla - \frac{2e}{\hbar c}\mathbf{A}\right)^2 + \frac{1}{\eta}\left(-\tau + \frac{2a}{Nd}|\psi_n|^2\right)\right]\psi_n \\
+ \tilde{b}[2\psi_n - \psi_{n+1}\exp(-i\chi_n) - \psi_{n-1}\exp(i\chi_n)] = 0, \\
j_z(n, n+1) = \frac{2e\tilde{b}d}{\hbar}[\psi_n^*\psi_{n+1}\exp(-i\chi_n) - \psi_n\psi_{n+1}^*\exp(i\chi_n)], \\
\mathbf{j}(n) = -\frac{ie\hbar}{m}(\psi_n^*\nabla\psi_n - \psi_n\nabla\psi_n^*) - \frac{4e^2}{mc}\mathbf{A}(n)|\psi_n|^2, \\
\chi_n = \frac{2ed}{\hbar c}A_z(n), \qquad \nabla = \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}\right), \qquad \mathbf{A} = (A_x, A_y), \\
\mathbf{j} = (j_x, j_y), \qquad \psi_n = \sqrt{\frac{7\zeta(3)N}{16(\pi T_c)^2}}\Delta_n,
\end{gathered} \tag{10}$$

where N is the electron density, $j_z(n, n+1)$ is the current density between the layers n and (n + 1), and $\mathbf{j}(n)$ is the current density in the layer n. The coefficients $\tilde{b}$ and η in (10) are determined by the expressions

$$\begin{gathered}
\tilde{b}_{(b)} = \frac{b^2}{\varepsilon_F}, \qquad \tilde{b}_{(h)} = \frac{\pi^3}{7\zeta(3)}\frac{\hbar D_\perp T_c}{d^2\varepsilon_F}, \\
\eta_{(c)} = \frac{7\zeta(3)}{8}\frac{\varepsilon_F}{(\pi T_c)^2}, \qquad \eta_{(d)} = \frac{\pi}{4}\frac{mD_{\|}}{\hbar T_c}.
\end{gathered} \tag{11}$$

In the derivation of (10) we used the condition $\tau \ll 1$, the condition (2) or (9), and assumed that $A_z(n)$ varies slowly with the coordinate n (this condition is satisfied at $H \ll \Phi_0/d^2$). Equations (10) contain a nonlinear dependence of j_z on the magnetic field, which is characteristic of the Josephson term. In the very narrow temperature interval $\tau \ll b^2/T_c^2$ it is possible to change over from these finite-difference equations to ordinary Ginzburg-Landau equations, which were indeed used in[1,2] to describe the magnetic properties of layered superconductors. In this case, however, the conduction between j_z and A_z is linear, and apart from the strong anisotropy the layered systems reveal no differences whatever from ordinary type-II superconductors.

VORTEX FILAMENTS IN PARALLEL FIELDS

In the temperature region $\tau \gg b^2/T_c^2$ and in not too strong fields $H_{\|}$, the influence of the field on the value of $|\psi_n|$ can be neglected, and we can regard $|\psi_n|$ as a constant. We then get from (10) and from Maxwell's equations for the field

$$\frac{\partial H}{\partial z} = -\frac{1}{\lambda_L^2}\left(\frac{\hbar c}{2e}\frac{\partial\varphi}{\partial x} - A_x\right), \qquad \lambda_L^2 = \frac{mc^2}{4\pi e^2 N_s(T)}, \tag{12a}$$

$$\frac{\partial H}{\partial x} = \frac{4\pi}{c}j_c\sin\frac{2ed}{\hbar c}\left(\frac{\hbar c}{2e}\frac{\partial\varphi}{\partial z} - A_z\right), \qquad j_c = e\tilde{b}d\frac{N_s(T)}{\hbar^3}. \tag{12b}$$

We note that for the band motion of the electron along the z axis ($l_\perp \gg d$), Eqs. (12) are valid in the entire temperature region $\tau \gg b^2/T_c^2$, since the equation for the current in the right-hand side of (12b) can be obtained in second-order perturbation theory from the Hamiltonian of the electron motion between layers (the tunnel Hamiltonian), inasmuch as the condition (2) is satisfied. Calculations analogous to those given in[6] for the Josephson junctions yield

$$\begin{gathered}
j_c = 4eb^2T\sum_{\omega,\mathbf{p}}F_1^+(\mathbf{p},\omega)F_2(\mathbf{p},-\omega) = \frac{1}{\pi}meb^2\frac{N_s(T)}{\hbar^3 N}, \\
\frac{N_s(T)}{N} = \pi T\Delta^2(T)\sum_\omega[\omega^2 + \Delta^2(T)]^{-3/2}, \qquad \frac{mb^2}{\pi N} = \tilde{b}_{(b)}d,
\end{gathered} \tag{13}$$

where F_1 and F_2 are the Gor'kov functions for the neighboring layers; the condition that $|\Delta_n|$ be constant is used. Apparently, Eqs. (12) are valid in the entire range of temperatures also for the hopping mechanism of an electron motion between layers; it is only necessary to replace in them $\tilde{b}$ by $\tilde{b}_{eff}(T)$.

Eliminating from (12) the phase φ and the vector potential A, we obtain an equation for the magnetic field H(x, z) in the vortex filament. The boundary condition for this equation requires that the total magnetic-field flux through the xz plane be equal to the flux quantum Φ_0 (it follows from the condition of the uniqueness of the phase on going over a closed contour around the vortex filament). In terms of the dimensionless variables u, v, and h given by

$$\begin{gathered}
u = x/\lambda_j, \quad v = z/\lambda_L, \quad h = H/H_0, \\
H_0 = \Phi_0/2\pi d\lambda_j, \quad \lambda_j^2 = c\Phi_0/8\pi^2 dj_c,
\end{gathered} \tag{14}$$

the equation and boundary conditions are

$$h = \frac{\partial^2 h}{\partial v^2} + \frac{\partial^2 h}{\partial u^2}\left[1 - \left(\frac{\partial h}{\partial u}\right)^2\right]^{-1/2}, \qquad \int h\,du\,dv = 2\pi\frac{a}{\lambda_L}. \tag{15}$$

The solution of (15) can be easily found by using the small parameter d/λ_L. In the region $\rho = \sqrt{u^2 + v^2} \gg d/\lambda_L$ we have $|\partial h/\partial u| \ll 1$, and the solution takes the form

$$h = (d/\lambda_L)K_0(\rho), \tag{16}$$

where $K_0(\rho)$ is a Bessel function of zero order of imaginary argument. In the region $\rho \lesssim d/\lambda_L \ll 1$ (in the region of the "core" of the vortex element), it is necessary to solve the nonlinear equation (15) for h. It follows from (15) that h, $\partial h/\partial u$, $\partial h/\partial v \sim 1$ inside the "core," therefore, just as in the case of an ordinary vortex filament, the "core" makes a small contribution to the total magnetic flux and to the vortex energy. From (16) we obtain for the energy $\mathscr{F}$ of the vortex filament and the field $H_{c1}(\|)$

$$\begin{gathered}
\mathscr{F} \approx \frac{H_0^2\lambda_L\lambda_j}{8\pi}\int du\,dv\left[\left(\frac{\partial h}{\partial u}\right)^2 + \left(\frac{\partial h}{\partial v}\right)^2 + h^2\right] = \frac{\Phi_0^2}{(4\pi)^2\lambda_L\lambda_j}\left(\ln\frac{\lambda_L}{d} + \varepsilon\right), \\
H_{c1}(\|) = \frac{\Phi_0}{4\pi\lambda_L\lambda_j}\left(\ln\frac{\lambda_L}{d} + \varepsilon\right),
\end{gathered} \tag{17}$$

where the quantity $\epsilon \sim 1$ is determined by the energy of the "core" of the filament. It is seen from (16) that in the region outside the "core" the vortex filament in a layered superconductor is similar to the ordinary filament of a strongly anisotropic type-II superconductor (see, e.g., Chap. 3 of[5]). The difference lies in the fact that in the layered compounds the superconducting state is not destroyed also in the region of the filament "core" (just as in the case of a vortex filament in Josephson junctions[6]).

It follows from (16) and (17) that to determine the vortex structure of layered superconductors in fields $H \ll \Phi_0\lambda_L/d^2\lambda_j$ and at temperatures (3) one can use the entire theory of the vortical state of type-II superconductors, by multiplying in the corresponding expressions the scales along the coordinate x by the factor λ_j/λ_L, and replacing ξ by d. In particular, the lowest energy is possessed by a triangular lattice, and for the moment M the dependence of the quantity $-4\pi M - H_{c1}(\parallel)$ on $H_{\parallel} - H_{c1}(\parallel)$ can be obtained from the calculations of[7], by reducing the scales of both quantities by a factor λ_j/λ_L. In fields $H \gtrsim \Phi_0\lambda_L/d^2\lambda_j$, the nonlinear "cores" of the filaments overlap and the approximation employed above no longer holds. At temperatures $\tau \ll b^2/T_c^2$, the vortex filament does not differ in any way, apart from the strong anisotropy, from the ordinary filament, and the filament "core" is in the normal state in this temperature region[2].

UPPER CRITICAL FIELD H_{c2} AND INHOMOGENEOUS STATE OF LAYERED SUPERCONDUCTORS

Near T_c, the field H_{c2} can be obtained with the aid of an equation of the Ginzburg-Landau type. We take the vector potential in the form $\mathbf{A} = (Hy \sin\theta, 0, -Hy \cos\theta)$, so that $H_{\parallel} = H\cos\theta$ and $H_{\perp} = H\sin\theta$. We can then assume that the order parameter depends only on y and, adding to (10) a term that takes into account the paramagnetic effect, we obtain equations for the determination of $H_{c2}(\parallel)$:

$$\eta\mathscr{E} + {}^7/_4\zeta(3)\ \gamma^2H^2/\pi^2T_c^2 = \tau,\ \gamma = {}^1/_2 g\mu_B, \tag{18}$$

$$\left\{-\frac{\hbar^2}{2m}\frac{d^2}{dy^2} + \frac{2e^2H^2\sin^2\theta}{mc^2}y^2 + 2\tilde{b}\left(1 - \cos\frac{2e\,dH\cos\theta}{c\hbar}y\right) - \mathscr{E}\right\}\psi = 0, \tag{19}$$

where $\mathscr{E}$ in (18) is the smallest eigenvalue of Eq. (19). Let us consider the case of a purely parallel field. Then (19) goes over into a Mathieu equation. At $\tau \ll b^2/T_c^2$, the field $H_{c2}(0)$ is small, and in the quasiclassical approximation we obtain for (19)

$$\mathscr{E} = edH(2\tilde{b}/mc^2)^{1/2}. \tag{20}$$

In this temperature region the paramagnetic effect can be neglected, and $H_{c2}(0)$ coincides with the usual expression of the Ginzburg-Landau theory of the anisotropic superconductor[1,2]. But at $\tau \gg b^2/T_c^2$ we have $\mathscr{E} \lesssim 2\tilde{b}$, $\eta\mathscr{E} \ll \tau$, and the field $H_{c2}(0)$ is determined by the paramagnetic effect:

$$H_{c2}(0) = \frac{2\pi}{\sqrt{7\zeta(3)}}\frac{T_c\sqrt{\tau}}{\gamma}. \tag{21}$$

Thus, at $\tau \gg b^2/T_c^2$ the field H_{c2} is determined only by the orbital motion of the electrons inside the layers in the field $H_{\perp}$ and by the paramagnetic effect. In the region $1 \gg \tau \gg b^2/T_c^2$ we obtain the field $H_{c2}(\theta)$ from (18) with $\mathscr{E} = eH\hbar\sin\theta/mc$.

At lower temperatures, in the case of layered "dirty" superconductors, we can determine $H_{c2}(\theta)$ by using the ordinary theory of "dirty" superconductors (it is only necessary to replace H in all the expressions that take into account the orbital effects by $H\sin\theta$). In particular, at low temperatures $T < 0.55T_c$, the inhomogeneous Larkin–Ovchinnikov–Fulde–Ferrell state[8,9] in "dirty" superconductors is apparently not realized[10], and the transition from the normal state to the BCS state becomes a first-order transition.

For $H_{c2}(\parallel)$ at $T = 0$ we obtain $H_p = \Delta(0)/\gamma\sqrt{2}$, where $\Delta(0)$ is the value of the gap at $T = 0$. We note that the the spin-orbit interaction, according to[11], suppresses the paramagnetic effect and consequently increases H_{c2}. This effect becomes most noticeably pronounced in a parallel field.

In pure layered superconductors at $T < 0.55T_c$, there can exist an inhomogeneous state. In layered superconductors, the conditions for its realization are more favorable than in isotropic superconductors, inasmuch as the orbital motion of the electrons can be neglected at small θ. Moreover, the region of existence of this state in layered superconductors is broader than in isotropic superconductors since, owing to the quasi-two-dimensional character of the system, the field of the transition from the normal state into the inhomogeneous state at $T = 0$ turns out to be $\sqrt{2}$ times larger than H_p (in the three-dimensional isotropic case it is equal to $1.07H_p$ without allowance for the orbital motion of the electrons).

The field of the transition from the normal state into the inhomogeneous state at $\theta = 0$ is determined by the largest value of H at which a solution exists for the equation

$$K_t(p, H, T) = 1. \tag{22}$$

The kernel $K_t(p, H, T)$ is obtained from (6) by replacing ω with $\omega - i\gamma H$:

$$K(p, H, T) = 4\lambda N(0)T\sum_{\omega}\int_{-pv_F}^{+pv_F}\frac{|\omega|\,d\Omega}{\sqrt{p^2v_F^2 - \Omega^2}\,[4\omega^2 + (2\gamma H + \Omega)^2]}. \tag{23}$$

At $T = 0$ we can use (22) and (23) to determine the dependence of $h = \gamma H$ on $P = pv_F/2$ by means of the expression

$$h^2 + |h^2 - P^2| + \sqrt{(h^2 + |h^2 - P^2|)^2 - P^4} = \Delta^2(0). \tag{24}$$

From (24) we see that the maximum value of h is reached at $P = \Delta(0)$, and is equal to $\Delta(0)$, so that at $T = 0$ we obtain $H_{c2}(\parallel) = \sqrt{2}H_p$.

The author thanks the participants of V. L. Ginzburg's seminar, especially A. I. Rusinov, for a useful discussion of the work.

Note added in proof (20 April 1972). A recent paper (R. C. Morris and R. V. Coleman, Phys. Rev. B7, 991, 1973) reports measurement of the anisotropy of the field H_{c2} in $TaS_2(Py)_{1/2}$ at $T = 2.84°K$ ($T_c = 3.25°K$). The results of these measurements are well described by formula (18) with $\mathscr{E} = H\tau\sin\theta/H_{c2}(\perp)$. For the ratio $H_{c2}^2(\theta)/H_{c2}^2(\perp)$, the experimentally obtained values were 335 and 530 at angles 3° and 2°, whereas the calculations yielded 315 and 560. From data on the resistance across the layers in $TaS_2(Py)_{1/2}$ (A. H. Thompson, F. R. Gamble, and R. F. Koehler, Jr., Phys. Rev. B5, 2811, 1972), condition (9) is satisfied for this compound, since $\hbar D_{\perp}/d^2 < 0.2°K$, and $TaS_2(Py)_{1/2}$ is indeed a superconductor with Josephson interaction of the layers.

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Translated by J. G. Adashko
238