Inhomogeneous state and the anisotropy of the upper critical field in layered superconductors with Josephson layer interaction

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Anisotropy of the upper critical field H_{c2} in layered superconductors with Josephson interaction of the layers is investigated. It is shown that in pure (along the layer) superconductors at low temperatures an inhomogeneous state is realized for field directions that are close to the layer direction, provided that the Cooper pairs in the field H_{c2} are not in the lower Landau orbit. The angular dependence of H_{c2} at $T = 0$ and the temperature dependence of H_{c2} (||) are obtained for pure superconductors. The dependence of H_{c2} (⊥) on temperature and purity of the crystals is investigated. It is shown that in dirty superconductors the transition from the normal to superconducting state at low temperatures may be a first-order transition for field directions close to a parallel direction and a second-order transition for other directions. The experimental data for $TaS_2(Py)_{1/2}$ are analyzed and it is shown that Josephson interaction of layers occurs in this compound.

1. INTRODUCTION

It has already been noted [1,2] that in the case of intercalation of layered compounds of the TaS_2 with molecules, superconductors with Josephson interaction between layers can be obtained. In the case of the hopping mechanism of conductivity between layers, an interaction of this type is realized if the following condition is satisfied:

$$\hbar / \tau_\perp \ll \Delta(T), \qquad (1)$$

where $\tau_\perp$ is the time between the hops of the electrons from one layer to the nearest neighboring one, and $\Delta(T)$ is the superconducting gap at the temperature T. Superconductors in which condition (1) is satisfied will be called layered superconductors with Josephson interaction of the layers (LSJI).

The value of the lower critical field $H_{c1}(\parallel)$ for the direction parallel to the layers and the structure of the vortical state in the LSJI were obtained earlier [1]. It is shown in the same reference that the anisotropy of H_{c1} and other features of the magnetic properties of LSJI in weak fields are due to the nonlinear dependence of the Josephson current between the layers on the vector potential **A**. In the present article we investigate the anisotropy of the upper critical field of LSJI. An important factor in the mechanism whereby the nuclei of the superconducting phase are produced is the quasi-two-dimensional character of the LSJI [3], which is also connected with the Josephson interaction of the layers. It was shown earlier [1] that if the condition (1) is satisfied near T_c (when $T_c - T \ll T_c$), then the field H_{c2} can be obtained without allowance for the motion of the electrons between the layers. It is clear that this conclusion is also valid in the region of lower temperatures. Therefore, if the condition (1) is satisfied, then the motion of the electrons in the LSJI can be regarded as two-dimensional in the self-consistent field approximation in calculation of the parameters that determine the appearance of the superconductivity. We note that by going outside the framework of the self-consistent-field approximation, i.e., by taking the phase fluctuations into account, we arrive, in a truly two-dimensional system, to a destruction of the superconducting long-range order. This does not occur for LSJI, since the phase fluctuations are suppressed by the Josephson interaction in the layers. Within the framework of the two-dimensional motion of the electrons the self-consistent-field approximation turns out to be valid because the phase fluctuations exert an essential influence on the superconducting properties only in a very close vicinity of the transition point, at practically arbitrarily small interaction between the layer [3-6].

For the case of a field parallel to the layers, in a pure superconductor (mean free path inside the layer $l \gg \xi_0$), the orbital motion of the electrons is inessential, and H_{c2} is determined only by the paramagnetic effect. Then, at temperatures $T < 0.55T_c$, an inhomogeneous state is realized [7,8], and at $T = 0$ we have

$$H_{c2}(\parallel) = \sqrt{2}H_p = \Delta(0)/\mu_0, \text{ where } \mu_0 = g\mu_B/2.$$

For all other field directions it is necessary when determining H_{c2} to take into account also the orbital effect connected with the two-dimensional motion of the electrons inside the layers.

We shall show below (see Sec. 3) that when account is taken of both the paramagnetic and the orbital effects, the inhomogeneous state is realized for field directions close to parallel, and in the LSJI it corresponds to the situation in which the Cooper pairs in the field H_{c2} are not on the lowest Landau orbit. In the same section, we obtain the angular dependence of H_{c2} at $T = 0$. The temperature dependence of $H_{c2}(\parallel)$ is investigated in Sec. 4. In Sec. 5 we obtain the dependence of $H_{c2}(\perp)$ on the temperature and the degree of the purity of the LSJI.

Section 6 is devoted to dirty LSJI. In them, the inhomogeneous state is not realized, and the transition from the normal state to the superconducting state may turn out to be a first-order transition for field directions close to parallel, and a second-order transition for other directions. In this section we take into account the influence of the spin-orbit scattering on the type of transition and on the value of H_{c2}. In the last section we analyze the experimental data for $TaS_2(Py)_{1/2}$ and show that this compound is of the LSJI type, and that an inhomogeneous state can apparently be observed in it.

2. EQUATION FOR THE DETERMINATION OF H_{c2}

If the condition (1) is satisfied, then in the calculation of H_{c2} we should take into account the paramagnetic effect and the orbital motion of the electrons inside the layers (in the x, y plane) in a field $H_Z = H \sin\theta$. Assuming the transition from the normal state to the superconducting state to be a second-order transition, we write down the linear equation for the order parameter

$$\Delta(\mathbf{r})\ln\frac{T_c}{T}=\int d^2\mathbf{r}' X(\mathbf{r},\mathbf{r}')\Delta(\mathbf{r}'),\qquad \mathbf{r}=(x,y), \tag{2}$$

from which H_{c2} is determined as the maximum value at which a nontrivial solution for $\Delta(\mathbf{r})$ exists [9].

To describe the orbital motion of the electrons in a field $H_Z = H\sin\theta$, we use a quasiclassical approximation, since we shall show below that the condition for the applicability of this approximation

$$e\hbar H_{c2}\sin\theta/mc \ll \pi T \tag{3}$$

is satisfied at practically all temperatures ($T \gg T_c^2/\epsilon_F$).

In the lowest approximation in the (nonmagnetic) impurity concentration, we obtain for the kernel of Eq. (2) [10]

$$\begin{gathered} X(\mathbf{r},\mathbf{r}')=\sum_\omega \frac{\pi T}{|\omega|}\delta^2(\mathbf{r}-\mathbf{r}')-S_\omega(\mathbf{r},\mathbf{r}'),\\ S_\omega(\mathbf{r},\mathbf{r}')=S_\omega^0(\mathbf{r},\mathbf{r}')+\frac{\hbar v_F}{2\pi T l}\int d^2\mathbf{r}'' S_\omega^0(\mathbf{r},\mathbf{r}'')S_\omega(\mathbf{r}'',\mathbf{r}'),\\ S_\omega^0(\mathbf{r},\mathbf{r}')=T(\rho\hbar v_F)^{-1}\exp\left\{\left[-2|\omega|+\frac{\hbar v_F}{l}+2i\mu_0 H\,\mathrm{sgn}\,\omega\right]\frac{\rho}{\hbar v_F}\right.\\ \left.+\frac{2ie}{\hbar c}\int_{\mathbf{r}}^{\mathbf{r}'}\mathbf{A}(\mathbf{s})\,d\mathbf{s}\right\},\qquad \boldsymbol{\rho}=\mathbf{r}-\mathbf{r}', \end{gathered} \tag{4}$$

where $\omega = (2k+1)\pi T$ and the two-dimensional vector potential $\mathbf{A} = (A_x, A_y)$ corresponds to the field $H_Z = H\sin\theta$.

In analogy with the isotropic three-dimensional case [10], it can be shown that $S_\omega^0(\mathbf{r},\mathbf{r}')$ is a function of only one operator $(i\nabla - 2ie\mathbf{A}/\hbar c)^2$, and consequently the solutions of equations (2) and (4) will be the wave functions of the electron motion in the field $H_Z = H\sin\theta$. Choosing $\mathbf{A}$ in the form $A_x = Hx\sin\theta$ and $A_y = 0$, we obtain

$$\begin{gathered} \int_{\mathbf{r}}^{\mathbf{r}'}\mathbf{A}(\mathbf{s})\,d\mathbf{s}=\frac{2ieH\sin\theta}{\hbar c}(x+x')(y-y'),\\ \Delta(\mathbf{r})=\exp\left\{\frac{2eH\sin\theta}{\hbar c}\left[ix_0y-\frac{1}{2}(x-x_0)^2\right]\right\}H_n\left[(x-x_0)\left(\frac{2eH\sin\theta}{\hbar c}\right)^{1/2}\right] \end{gathered} \tag{5}$$

where $H_n(x)$ are Hermite polynomials and the solutions (5) are degenerate with respect to the parameter x_0.

Substitution of (5) in (2) and (4) yields

$$\begin{gathered} \ln\frac{T_c}{T}=\sum_\omega \frac{\pi T}{|\omega|}-\frac{s_\omega}{1-\hbar s_\omega/2\pi T\tau},\\ s_\omega=\frac{T(-1)^n}{\hbar v_F}\int\frac{d^2\rho}{\rho}\exp\left[\left(-2|\omega|+\frac{\hbar}{\tau}-2i\mu_0 H\,\mathrm{sgn}\,\omega\right)\frac{\rho}{\hbar v_F}\right.\\ \left.-\frac{eH\sin\theta}{2\hbar c}\rho^2\right]L_n\left(\frac{eH\sin\theta}{\hbar c}\rho^2\right), \end{gathered} \tag{6}$$

where $L_n(x)$ are Laguerre polynomials and $\tau = l/v_F$.

In the case of pure superconductors, using the digamma function $\psi(x)$ and changing over to the momentum presentation $\mathscr{P}$ with respect to the variable $\boldsymbol{\rho}$, we obtain from (6)

$$\begin{gathered} \ln\frac{T_c}{T}=\frac{\hbar c(-1)^n}{\pi eH\sin\theta}\int_0^\infty \mathscr{P}d\mathscr{P}\exp\left(-\frac{\mathscr{P}^2\hbar c}{2eH\sin\theta}\right)L_n\left(\frac{\mathscr{P}^2\hbar c}{eH\sin\theta}\right)\cdot\\ \times\,\mathrm{Re}\int_{-\mathscr{P}v_F}^{+\mathscr{P}v_F}\frac{d\Omega}{(\mathscr{P}^2v_F^2-\Omega^2)^{1/2}}\left\{\psi\left(\frac{1}{2}+i\frac{\Omega+2\mu_0 H}{4\pi T}\right)-\psi\left(\frac{1}{2}\right)\right\}. \end{gathered} \tag{7}$$

3. INHOMOGENEOUS STATE AND ANISOTROPY OF H_{c2} IN PURE LSJI AT ZERO TEMPERATURE

To change over to the temperature $T = 0$, we use the asymptotic digamma function. Then, after integrating with respect to Ω, Eq. (7) takes the form

$$\begin{gathered} (-1)^n\ln h=\int_0^{\alpha h}dx\,e^{-x}L_n(2x)\ln\left[1+\left(1-\frac{x}{\alpha h}\right)^{1/2}\right]\\ -\frac{1}{2}\int_{\alpha h}^\infty dx\,e^{-x}L_n(2x)\ln\left(\frac{x}{\alpha h}\right), \end{gathered} \tag{8}$$

$$h=\frac{\mu_0 H}{\Delta(0)},\qquad \alpha=\frac{H_{c20}}{H_p\gamma\sqrt{2}\sin\theta},\qquad H_{c20}=\frac{2c\Delta^2(0)}{e\hbar v_F^2}, \tag{9}$$

where $\ln\gamma = C$ is the Euler constant.

Equation (8) at a given α determines the dependence of h on the number of the Landau orbit n, and the field H_{c2} corresponds to the maximum of the function h_n with respect to the discrete variable n. At small α, the maximum of h_n is reached at $n = 0$, and the dependence $h_{c2}(\alpha)$ at $\alpha \ll 1$ can be obtained from the equation

$$\ln(h/\alpha)+(2\ln 2+1)ch-C=0. \tag{10}$$

From (10) we obtain at $H_{c20} \ll H_p$ the value $H_{c2}(\perp) = H_{c20}$.

The numerically obtained plots of $h_n(\alpha)$ at $\alpha > 0.5$ for n from 0 to 6 are shown in Fig. 1, from which it is seen that at $\alpha > 1.25$ the solutions for $\Delta(\mathbf{r})$ corresponding to H_{c2} are functions of excited Landau orbits. The solid curve in Fig. 1 corresponds to the maximum (at a given α) value of h_n and it yields the function $h_{c2}(\alpha)$. As $\alpha \to \infty$, we have $h_{c2} \to 1$ in accordance with the results obtained in [1], and according to the data for $n \le 12$ the value n_{max}, which determines h_{c2}, increases linearly with increasing α, or, more accurately speaking, n_{max} coincides approximately with the integer part of the quantity $\alpha/1.8$. The function $H_{c2}(\theta)$ can now be obtained from (9), (10), and Fig. 1. The inhomogeneous state ($n \ge 1$) is realized at an angle $\theta < \theta_c = \sin^{-1}(H_{c20}/3.14H_p)$.

Thus, whereas in a three-dimensional isotropic superconductor the inhomogeneous state can be realized only at sufficiently large value of H_{c20}/H_p ($\gtrsim 2$)[11], in LSJI the inhomogeneous state is realized at small values of H_{c20}/H_p, but in a narrow angle interval $\theta \lesssim H_{c20}/H_p$. The dependence of H_{c2} on θ in LSJI is characterized by two features connected with the realization of the inhomogeneous state, namely the sharp increase of H_{c2} (by an approximate factor 1.5) when θ decreases from θ_c to zero, and a nonmonotonic dependence of H_{c2} on θ at $\theta < \theta_c$. The presence of weak oscillations in the plot

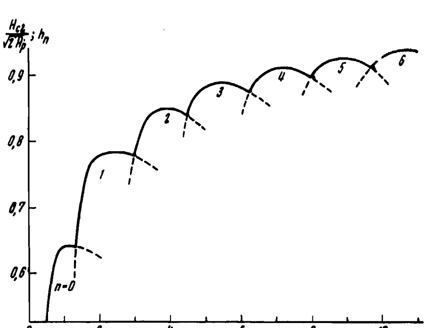

FIG. 1. Plots of $H_{c2}/\sqrt{2}H_p$ and h_n against the parameter α at zero temperature

of H_{C2} against θ is due to the fact that when the angle θ decreases from θ_C to zero a transition to higher Landau orbits takes place in the system (see Fig. 1). However, the appearance of oscillations in the plot of H_{C2} against θ can apparently be observed only in superconductors with not too small values of H_{C20}/H_p.

We can now justify the validity of the quasiclassical approximation used above. Since the paramagnetic effect only decreases H_{C2}, it follows that $H_{C2} < H_{C20}/\sin\theta$ and at $T \gg T_C^2/\epsilon_F$ the condition (3) is indeed satisfied.

4. THE TEMPERATURE DEPENDENCE OF $H_{c2}(\parallel)$ IN PURE LSJI

For a field parallel to the layers ($\sin\theta \to 0$), the dependence of $\Delta(\mathbf{r})$ on the coordinates is of the form $\exp(i\mathscr{P}x)$, and Eq. (7) will go over into

$$\ln\frac{T_c}{T} = \frac{1}{\pi}\int_{-\mathscr{P}v_F}^{+\mathscr{P}v_F}\frac{d\Omega}{(\mathscr{P}^2v_F^2-\Omega^2)^{1/2}}\left\{\psi\left(\frac{1}{2}+i\frac{2\mu_0H+\Omega}{4\pi T}\right)-\psi\left(\frac{1}{2}\right)\right\}. \quad (11)$$

Equation (11) determines the dependence of H on $\mathscr{P}$, and $H_{C2}(\parallel)$ corresponds to the maximum of this function with respect to the variable $\mathscr{P}$.

Let us find first the asymptotic form of $H_{C2}(\parallel)$ at $T \ll T_C$. We introduce the dimensionless variables $\omega = \Omega/2\Delta(0)$, $p = \mathscr{P}v_F/2\Delta(0)$, and $\tilde{t} = \pi T/\Delta(0)$. The dependence of h on p at $T = 0$ was obtained earlier [1], and is determined from the equation

$$h^2 + |h^2 - p^2| = {}^1/_2(1+p^4). \quad (12)$$

The maximum of h is reached at $p = 1$, and, as seen from (12), the function h(p) is not analytic in the vicinity of this point. One can therefore expect the dependence of $h_{C2}(\parallel)$ on T to be likewise nonanalytic as $T \to 0$. We use the following integral representation for the digamma function [12]

$$\psi(x) = \ln x - \frac{1}{2x} - 2\int_0^\infty\frac{y\,dy}{(y^2+x^2)(e^{2\pi y}-1)}. \quad (13)$$

After substituting (13) in (11), we obtain an equation that determines the dependence of h on p and $\tilde{t}$:

$$f(h,p) = \frac{\tilde{t}^2}{\pi}\int_{-1}^{+1}\frac{d\omega}{(1-\omega^2)^{1/2}[\tilde{t}^2+(h+p\omega)^2]} - \frac{1}{2\pi}\int_0^{t^2}dx \times$$
$$\times\int_{-1}^{+1}\frac{d\omega}{(1-\omega^2)^{1/2}[x^2+(h+p\omega)^2]} + \frac{2}{\pi}\mathrm{Re}\int_{-1}^{+1}\frac{d\omega}{(1-\omega^2)^{1/2}}\int_0^\infty\frac{x\,dx\,(e^{2\pi x}-1)^{-1}}{x^2+[{}^1/_2+i(h+p\omega)/2\tilde{t}]^2},$$

$$f(h,p) = \frac{1}{\pi}\int_{-1}^{+1}\frac{d\omega}{(1-\omega^2)^{1/2}}\ln|2h+2p\omega| = \begin{cases}\ln[h+(h^2-p^2)^{1/2}], & h \geqslant p,\\ \ln p, & p > h.\end{cases} \quad (14)$$

The integration with respect to ω in (14) can be carried out exactly; however, the expressions obtained after integration are cumbersome, and we shall not present them here. An analysis of these expressions shows that at $\tilde{t} \ll 1$ the dependence of h on p in the hp plane intersects the line $h = p$ and continues to move upward, i.e., $\partial h/\partial p > 0$ at $|h - p| \ll \tilde{t} \ll 1$. Therefore the maximum of h with respect to the variable p is reached at $p > h$ either in the region $p - h \sim \tilde{t}$, or in the region $p - n \gg \tilde{t}$. In the former case $1 - h \sim \tilde{t}^{1/2}$, and in the latter case higher values of h are reached. Indeed, at $p - h \equiv z \gg \tilde{t}$ we obtain

$$h(z,\tilde{t}) = 1 - z - \frac{\sqrt{2}}{12}\tilde{t}^3 z^{-5/2}. \quad (15)$$

Finding the maximum of h(z, t) with respect to the variable z, we obtain at $\tilde{t} \ll 1$

$$z_{max} = \left(\frac{5\sqrt{2}}{24}\tilde{t}^3\right)^{2/7}, \quad p_{max} = 1 - \frac{\sqrt{2}}{12}\left(\frac{5\sqrt{2}}{24}\right)^{-5/7}\tilde{t}^{6/7},$$
$$h_{c2}(\parallel) = h_{max} = 1 - \frac{7\sqrt{2}}{24}\left(\frac{5\sqrt{2}}{24}\right)^{-5/7}\tilde{t}^{6/7}. \quad (16)$$

It is seen from (16) that $z_{max} \gg \tilde{t}$ at $\tilde{t} \ll 1$ in accordance with the assumption made above. The condition $z_{max} \gg \tilde{t}$ is satisfied more accurately at $(288\,\tilde{t}/25)^{1/7} \ll 1$, and in this temperature region the asymptotic form (16) is valid.

The dependence of $h_{C2}(\parallel)$ on $t = T/T_C$ was investigated earlier [1] for t close to unity. In the region of intermediate values of t, the quantity $h_{C2}(\parallel)$ can be obtained only numerically. (To carry out numerical calculations it is more convenient to use in place of Eq. (11) with the digamma function an equation with summation over frequencies, and to integrate with respect to $\mathscr{P}$.) The dependence of $H_{C2}(\parallel)$ on t is shown in Fig. 2, which also shows the temperature dependence of the parameter p of the inhomogeneity of the state. From Fig. 2 it is seen that, in agreement with the results of [3], the inhomogeneous state is realized at $T < 0.55T_C$.

5. DEPENDENCE OF $H_{c2}(\perp)$ ON THE TEMPERATURE AND ON THE DEGREE OF PURITY OF THE CRYSTALS

Usually $H_{C2}(\perp) \ll H_p$, and in this case the paramagnetic effect can be neglected when $H_{C2}(\parallel)$ is calculated. We then obtain from (6) at $n = 0$

$$\ln\frac{1}{t} = \sum_{k=-\infty}^{+\infty}\left[\frac{1}{|2k+1|} - \frac{t\tilde{h}^{-1/2}\varphi(a)}{1-\lambda\tilde{h}^{-1/2}\varphi(a)}\right], \quad t = \frac{T}{T_c},$$
$$a = \frac{t\,|2k+1|+\lambda}{\tilde{h}^{1/2}}, \quad \lambda = \frac{\hbar}{2\pi T_c\tau}, \quad \tilde{h} = \frac{e\hbar v_F^2 H_{c2}(\perp)}{2\pi^2 c T_c^2}, \quad (17)$$
$$\varphi(a) = \sqrt{\pi}\,e^{a^2}\left(1 - \frac{2}{\sqrt{\pi}}\int_0^a e^{-x^2}dx\right).$$

Like Helfand and Werthamer [10], we plot $h^* = \tilde{h}/(-d\tilde{h}/dt)_{t\to1}$ against t, where $(-d\tilde{h}/dt)_{t\to1}$ is the derivative of $\tilde{h}$ with respect to t in the temperature region where the interaction of the layers still remains of the Josephson type, i.e., in the region $1 \gg 1 - t \gg \hbar/\tau_\perp T_C$. Owing to the quasi-two-dimensional character, we have

$$\left(\frac{d\tilde{h}}{dt}\right)_{t\to1} = 2\lambda^2\left[\frac{\pi^2\lambda}{4} - \psi\left(\frac{1}{2}+\frac{\lambda}{2}\right) + \psi\left(\frac{1}{2}\right)\right]^{-1}. \quad (18)$$

The results of the numerical calculation are shown in Fig. 3, from which it is seen that $h^*(t)$ in LSJI is lower the smaller λ. In the isotropic three-dimensional case [10] the situation is reversed, and $h^*(t)$ is smaller the larger λ. At $T = 0$, with accuracy no worse than 3%, the plot of $H_{C2}(\perp)$ against λ is approximated by the formula

$$H_{c2}(\perp) = H_{c20}(1 + 1{,}1\lambda) \quad (19)$$

for $\lambda \leq 6$.

6. TYPE OF PHASE TRANSITION AT H_{c2} IN DIRTY LSJI

We consider first the case of dirty LSJI ($l \ll \xi_0$) without allowance for spin-orbit scattering. We can then show that the inhomogeneous state is not realized in the limit of dirty LSJI. Indeed, if we wish to regard the transition from the normal state into the superconduct-

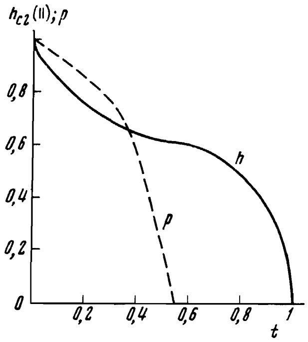

FIG. 2

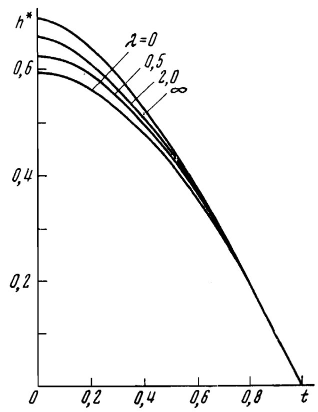

FIG. 3

FIG. 2. Dependence of $h_{C2}(\parallel)$ and of the inhomogeneity parameter p on the temperature.

FIG. 3. Dependence of h* on the temperature and on the mean free path.

ing state as a second-order transition, then we obtain for the determination of H_{C2} the equation [13]

$$\left\{\ln\frac{1}{t}-\operatorname{Re}\psi\left[\frac{1}{2}+\frac{D}{2\pi T}\left(\frac{d^2}{dx^2}-\frac{4e^2}{c^2}H^2x^2\sin^2\theta\right)+\frac{i\mu_0 H}{2\pi T}\right]+\psi\left(\frac{1}{2}\right)\right\}\Delta(x)=0, \tag{20}$$

where $D = v_F l/2$ is the coefficient of diffusion inside the layer. Taking for $\Delta(x)$ the solution corresponding to the Landau orbit with quantum number n, we obtain from (20)

$$\ln\frac{1}{t}=\operatorname{Re}\psi\left[\frac{1}{2}+\left(n+\frac{1}{2}\right)\frac{DeH\sin\theta}{\pi T}+\frac{i\mu_0 H}{2\pi T}\right]-\psi\left(\frac{1}{2}\right). \tag{21}$$

It is seen from (21) that the maximum field is reached at $n = 0$, and that the inhomogeneous state is impossible in dirty LSJI. It is clear that this conclusion does not change if we also take the spin-orbit scattering into account.

In accordance with Maki's results [14], the transition from the normal state to the superconducting state at small θ and at low temperatures may turn out to be of first order. In the analysis of the situation, we take into account also the spin-orbit scattering, in analogy with the procedure used for an isotropic three-dimensional superconductor by Werthamer, Helfand, and Hohenberg [15]. In dirty LSJI with a spin-orbit scattering time $\tau_2 \gg \tau$, we obtain for H_{C2} the equation

$$\ln\frac{1}{t}=\sum_{k=-\infty}^{+\infty}\left\{\frac{1}{|2k+1|}-\left[|2k+1|+\frac{\bar h}{t}+\frac{(\alpha\bar h/t)^2}{|2k+1|+(\bar h+\lambda_{so})/t}\right]^{-1}\right\}, \tag{22}$$

$$\bar h=\frac{eDH_{c2}\sin\theta}{\pi cT_c},\quad \lambda_{so}=\frac{1}{\pi T_c\tau_2},\quad \alpha=\frac{\mu_0 c}{eD\sin\theta}. \tag{23}$$

Equation (22) coincides fully with Eq. (28) from [15], except that the expressions for $\bar h$, α, and λ_{so} are different. An analysis of (22) at $T = 0$ [15] shows that at $\lambda_{so} < \lambda_{so}^{c} = 0.5139$ the transition from the normal state into the superconducting state turns out to be a second-order transition at $\alpha < \alpha_c$ and a first-order transition at $\alpha > \alpha_c$, where

$$\alpha_c=[1+1{,}589\lambda_{so}/\lambda_{so}^c]/[1-\lambda_{so}/\lambda_{so}^c]. \tag{24}$$

At $\alpha = \alpha_c$ we obtain $H_{C2}^{c} = H_p(0.5 + 0.795\lambda_{so}/\lambda_{so}^{c})$, and for smaller α we obtain the field $H_c^2 < H_{C2}^{c}$. At $\lambda_{so} \geq \lambda_{so}^{c}$, the transition to the superconducting state is a second-order transition regardless of the value of α and $H_{C2} \geq H_p$ as $\sin\theta \to 0$.

The case of nonzero temperatures at $\lambda_{so} = 0$ was considered by St. James et al. [13], and the results obtained there make it possible to draw the boundary between the regions of the first- and second-order transitions on the θ, T plane. Thus, in dirty LSJI with not too strong a spin-orbit scattering one can observe, at low temperatures, the replacement of a second-order transition by a first-order transition as the direction of the magnetic field approaches the parallel direction. For small t and λ_{so}, this replacement occurs at an angle $\sin\theta \approx \mu_0 c/eD$.

7. ANALYSIS OF EXPERIMENTAL DATA FOR $TaS_2(Py)_{1/2}$

From measurements of the electronic specific heat at low-temperatures [16] we can obtain the density of states, since the term linear in the temperature in the specific heat β is connected with the density of states $\overline{N(0)}$ by the relation

$$\beta={}^2/{}_3\pi^2\overline{N(0)}\,k_B{}^2, \tag{25}$$

where k_B is Boltzmann's constant. The quantity $\beta = (8.9 \pm 0.04)$ mJ/mole°K^2 yields $\overline{N(0)} \approx 10^{34}$ erg^{-1} cm^{-3} (molecular weight 285, density 4.1 g/cm^3 [7]). Now, knowing the lower bound of the resistivity $\rho_\perp$ across the layers [8], we can obtain an upper bound of $\hbar/\tau_\perp$, using the relation

$$1/D_\perp=\tau_\perp/d^2=2e^2\rho_\perp\overline{N(0)}, \tag{26}$$

where d is the distance between layers, equal to 12 Å for $TaS_2(Py)_{1/2}$. From $\rho_\perp > 6\Omega$-cm (at temperatures below 10°K) we obtain $\hbar/\tau_\perp < 0.17$°K. Comparing this estimate with the value $T_c = 3.25$°K, we verify that the condition (1) is satisfied in $TaS_2(Py)_{1/2}$ at least for temperatures $1 - T/T_c > 3\times10^{-4}$ and, consequently, the interaction of the layers is of the Josephson type at practically all the temperatures.

We note that the value of $\hbar/\tau_\perp$ can be obtained by measuring the field $H_{c1}(\parallel)$, for in accordance with the results of [1] we have

$$H_{c1}(\parallel)=\frac{c\hbar}{4e\lambda_L\lambda_j}\ln\frac{\lambda_L}{d},\quad \lambda_L{}^2=\frac{mc^2}{4\pi N_s e^2},\quad \lambda_j{}^2=\frac{7\zeta(3)c^2\hbar^3\tau_\perp}{8\pi^3me^2T_cd}, \tag{27}$$

where $\zeta(x)$ is the Riemann function and λ_L and λ_j are the respective depths of penetration of the field perpendicular and parallel to the layers.

The values of the coherence length ξ_0 and the mean free path l inside the layer can be obtained if we know (in addition to $\overline{N(0)}$ and T_c) the resistance $\rho_\parallel$ along the layers and the critical field $H_c(\perp)$. When ξ_0 and l are determined from the equations

$$l\xi_0=\frac{\hbar\gamma}{\pi^2e^2\rho_\parallel\overline{N(0)}\,T_c},\quad \lambda\Big/\left(-\frac{d\bar h}{dt}\right)_{t\to1}=\frac{\pi h^*e\rho_\parallel\overline{N(0)}\,T_c}{H_{c2}(\perp)}, \tag{28}$$

and the quantities λ, h*, and $(-d\bar h/dt)_{t\to1}$ are defined in Sec. 5.

The measured values of $\rho_\parallel$ and $H_{C2}(\perp)$ for $TaS_2(Py)_{1/2}$ are given by Morris and Coleman in [17]. Unfortunately, during the course of the measurements with one and the same crystal, the value of $\rho_\parallel$ varied in the course of time from 10^{-5} Ω-cm at the start of the experiment to 6×10^{-5} Ω-cm at the end. Apparently, the measurements of $H_{c2}(\perp)$ at $T = 2°K$ are among the earliest ones, since they yield the largest value of l. We can therefore take for the crystal at the start of the experiments $H_{c2}(\perp) = 1.4$ kOe and $\rho_{\parallel} = 10^{-5}$ Ω-cm at $T = 2°K$. In this case we obtain with the aid of (28) $\xi_0 = 4.4 \times 10^{-6}$ cm, $l = 3.5 \times 10^{-6}$ cm, and $v_F = 1.1 \times 10^7$ cm/sec. Inasmuch as in the quasi-two-dimensional case[1)] we have $\overline{N(0)} = m/2\pi d$, we get for the effective mass the value $m = 9 \times 10^{-27}$ g and $k_F = 10^8$ cm^{-1}.

The carrier density n is connected with k_F by the relation $n = k_F^2/2\pi d$, and for $TaS_2(Py)_{1/2}$ we obtain $n = 10^{22}$ cm^{-3}. If each Ta atom gives one electron to the conduction band, then the electron density should be of the order of 0.85×10^{22} cm^{-3}. According to the results of Thompson, Gamble, and Koehler [18], the carrier density in TaS_2 at $T < 20°K$ is about 1.5×10^{22} cm^{-3}. In $TaS_2(Py)_{1/2}$, the concentration of the conducting electrons should be somewhat more than half of this value, since the volume per Ta atom in this material is twice as large as in TaS_2 and since, according to the results of [19], pyridine (Py) adds approximately an additional 0.25 electron to the conduction band, increasing the carrier density by approximately 0.12×10^{22} cm^{-3}. The good agreement of all these estimates for n shows that the values of ξ_0 and l presented above are not far from the real ones.

Thus, in the study of Morris and Coleman [17], the $TaS_2(Py)_{1/2}$ crystal was of intermediate purity at the start of the measurements ($\lambda = 1.1$), but by the end of the experiments it must be regarded already as a dirty superconductor ($\lambda \approx 4$-6). Since the spin-orbit scattering is weaker by at least 2 orders of magnitude than the usual scattering, it follows that $\lambda_{so} < 0.05$, and the spin-orbit scattering for the investigated samples is immaterial. Therefore, when the field direction approaches parallel ($\theta \lesssim 2°$) and at temperatures $T \ll T_c$, there should be realized below H_{c2}, depending on the degree of purity of the crystal, either an inhomogeneous state or else a first-order transition from the normal state to the superconducting state [20,21].

Morris and Coleman [17] measured the anisotropy of H_{c2} at 2.84 and 1.4°K. In the former case the temperature is close to T_c ($t = 0.88$), the transition from the normal state to the superconducting state is a second-order transition, and in accordance with the results of [1] the dependence of H_{c2} on θ is given by the expression

$$\frac{H_{c2}^2(\theta)}{H_{c2}^2(\perp)} = \frac{1 + 2x^2 - (1 + 4x^2)^{1/2}}{2x^4 \sin^2\theta}, \quad x = \frac{H_{c2}(\perp)}{H_{c2}(\parallel)\sin\theta}, \qquad H_{c2}(\parallel) = \frac{2\pi T_c (1-t)^{1/2}}{\mu_0 \sqrt{7\zeta(3)}}. \tag{29}$$

At $T = 2.84°K$, the value of H_{c2} at $\theta = 4°$ is ~7 kOe, and $H_{c2}(\perp) = 0.5$ kOe (according to the results of Sec. 5, the value $H_{c2}(\perp) = 1.4$ kOe at $T = 2°K$ corresponds to a field $H_{c2}(\perp) = 0.45$ kOe at $T = 2.84°K$). From (29) we obtain $H_{c2}(\parallel) = 36.2$ kOe, and for the angles $\theta \geq 4°$ we have $x \ll 1$ and $H_{c2}^2(\theta)/H_{c2}^2(\perp) \approx 1/\sin^2\theta$. At $\theta = 3$ and 2° we obtain from (29) for the same ratio the values 315 and 560, respectively, whereas the experimental values are 335 and 530.

Thus, the agreement between the theoretical results [1] and the experimental data [17] for $T = 2.84°K$ is good, and confirms the premise that the interaction of the layers in $TaS_2(Py)_{1/2}$ is of the Josephson type. We note that Morris and Coleman [17] have observed a very slow approach of the resistance to the normal value when the field H was increased above H_{c2} at small θ and at $T = 2.84°K$. This variation of the resistance with the field is apparently connected with the superconducting fluctuations above H_{c2}. The paramagnetic effect facilitates the appearance (owing to the fluctuations) of superconducting regions with characteristic dimensions on the order of $\hbar v_F/\mu_0 H$ (it is precisely for this reason that an inhomogeneous state is realized in pure LSJI at $T < 0.55T_c$ and small θ). Therefore the influence of the fluctuations on the conductivity above H_{c2} (but below T_c) should be stronger than above T_c.

We consider now the data for $H_{c2}(\theta)$ at $T = 1.4°K$. At this temperature we have $H_{c2}(\perp) = 4.9$ kOe, and for this value of the field we obtain $l = 10^{-6}$ cm. Obviously, at such a small mean free path and at a temperature $T = 0.43T_c$ the inhomogeneous state is not realized. We apply the theory of dirty LSJI to this case. Then, according to the results of Sec. 6 and the book by St. James et al.[13], for angles $\theta \gtrsim 2°$ the transition from the normal state to the superconducting state is a second-order transition. The calculated theoretical values of $H_{c2}^2(\theta)/H_{c2}^2(\perp)$ for the angles $\theta \leq 6°$ turn out to be in this case lower than the experimental ones (they differ by factors 3.3, 2.5, and 1.5 for the respective angles 2°, 4°, and 6°). This difference can apparently be attributed to the uncertainty in the experimental determination of $H_{c2}(\theta)$ at small θ, owing to the weak and nonmonotonic growth of the resistance with increasing magnetic field above H_{c2}. At $\theta = 0$ and $T = 1.4°K$, the transition to the superconducting state from the normal state should be a first-order transition and should occur in a field $\sim 0.9H_p = 54$ kOe. Yet experimentally, at $\theta = 0$, the resistance amounts to only a small fraction of the resistance in the normal state, even in fields that are 2.5 times larger than $H_p = 60$ kOe. This effect is possibly connected with the realization of a metastable state. At $T = 0$, the metastable superconducting state with a homogeneous order parameter can be preserved in fields $H \leq \sqrt{2}H_p$ [22]. The region of metastable states with an inhomogeneous order parameter is apparently even broader.

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[1)]We note that according to [1] the critical temperature T_c in LSJI is determined by the density of states $N(0) = m/2\pi a$, where a is the thickness of the conducting layer.

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