

Diamagnetic fluctuations in layer superconductors and small superconducting particles

L. N. Bulaevskii

P. N. Lebedev Physics Institute, USSR Academy of Sciences

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Fluctuations of the diamagnetic moment in dirty layer superconductors with Josephson interaction between layers [of the $\text{TaS}_2(\text{Py})_{1/2}$ type] are investigated. The fluctuations are of a two-dimensional nature, not too close to the vicinity of the transition point. The contribution of thermal fluctuations and zero oscillations of Cooper pairs is evaluated. Above T_c , the contribution of zero oscillations, as contrasted to that of thermal fluctuations, decreases with the temperature and field strength, but very slowly; it is still appreciable at temperatures $T > T_c$ and field strengths $H > H_{c2}(0)$, provided that the collision time τ is sufficiently small. The fluctuation moment is calculated for a temperature $T = 0$ and fields $H > H_{c2}(0)$. The moment is determined only by zero oscillations of Cooper pairs. With increase of the field to $H = H_{c2}(0)$ from above, the fluctuation moment grows logarithmically and its decrease with increase in the field above $H_{c2}(0)$ is very slow. Similar results have also been obtained for the diamagnetic fluctuations of small superconducting particles.

1. INTRODUCTION

It follows from the papers of Lee and Payne,^[1] Kurkijarvi, Ambegaokar and Eilenberger,^[2] and Maki^[3] that the contribution to the diamagnetic moment above the superconducting transition point in dirty superconductors is determined not only by thermal fluctuations, but also by the zero-point oscillations of Cooper pairs. The contribution of the zero-point oscillations falls off with increasing magnetic field H and temperature $T - T_c$ much more slowly than the contribution of thermal fluctuations, since the characteristic parameters of the field and energy for the zero-point oscillations are the quantities $H_S = \Phi_0/l^2$ and \hbar/τ , respectively ($\Phi_0 = \pi\hbar c/e$ is the magnetic flux quantum, l and τ are the free-path length and time) and not $H_{c2}(0)$ and T_c , as is the case for thermal fluctuations. However, in three-dimensional superconductors, this contribution is nonsingular as $T \rightarrow T_c$ (or $H \rightarrow H_{c2}$) and is comparable in absolute magnitude with the Landau diamagnetism for conduction electrons in the normal state. Therefore, in three-dimensional superconductors, the contribution of the zero-point oscillations of the Cooper pairs to the diamagnetic moment is difficult to distinguish from the usual diamagnetism. In this paper, it will be shown that the situation is more encouraging in the two-dimensional and zero-dimensional cases, since the contribution of the zero point oscillations to the diamagnetic moment turns out to be large in comparison with the diamagnetism of the normal electrons, and it increases logarithmically with approach of the temperature T to T_c or of the field H to H_{c2} .

It was shown in^[4] that layer superconductors with Josephson tunneling between the layers behave like two-dimensional systems at temperatures that are not very close to T_c (or fields H not too close to H_{c2}). According to estimates,^[4] Josephson tunneling between layers is achieved in the intercalated compound of TaS_2 with pyridine ($\text{TaS}_2(\text{Py})_{1/2}$), and the majority of investigated samples of this compound are dirty superconductors, since the free path length l inside the layer is much less than the coherence length $\xi_0 = \hbar v_F/\pi\Delta_0$ (for the samples studied by Morris and Coleman,^[5] the ratio ξ_0/l varied approximately from 1 to 6). Therefore, $\text{TaS}_2(\text{Py})_{1/2}$ is an ideal object for investigation of the contribution of the zero-point oscillations of Cooper pairs to the diamag-

netic moment above the superconducting transition point. Evidently the most favorable conditions for such a study are achieved at low temperatures $T \ll T_c$ and fields $H \gtrsim H_{c2}(0)$, when the fluctuation diamagnetic moment is determined only by the zero-point oscillations.

2. INITIAL EQUATIONS AND TWO-DIMENSIONALITY OF FLUCTUATIONS IN LAYER SUPERCONDUCTORS WITH JOSEPHSON TUNNELING BETWEEN THE LAYERS

The free energy associated with the superconducting fluctuations is determined in the presence of a magnetic field by the expression^[6]

$$F = \frac{2}{\pi} \text{Sp} \int_0^\infty d\omega \left[\frac{1}{2} + \frac{1}{e^{\beta\omega} - 1} \right] \text{Im} \ln \mathcal{D}(\omega, A), \quad (1)$$

where $\beta = 1/T$ and \mathcal{D} is the propagation function of the fluctuation field of the Cooper pairs. It follows from^[4,6] that this function has the following form for a dirty layer superconductor with Josephson tunneling, with account of the paramagnetic effect:

$$\begin{aligned} \mathcal{D}(\omega, A) = & \lambda^{-1} \left[\frac{1}{2} \psi \left(\frac{1}{2} + \frac{\mathcal{E}_+}{4\pi T} \right) \right. \\ & \left. + \frac{1}{2} \psi \left(\frac{1}{2} + \frac{\mathcal{E}_-}{4\pi T} \right) - \psi \left(\frac{1}{2} \right) + \ln \frac{T}{T_c} \right]^{-1}, \quad (2) \\ \mathcal{E}_\pm(\omega, A) = & i\omega \pm 2i\mu H - \hbar D_{\parallel} \left(\frac{\partial}{\partial \mathbf{r}_{\parallel}} - \frac{2ie}{\hbar c} A_{\parallel} \right)^2 \\ & - \frac{\hbar D_{\perp}}{d^2} \left[1 - \cos \left(d \frac{\partial}{\partial z} - \frac{2ied}{\hbar c} A_z \right) \right], \quad (3) \end{aligned}$$

where $\psi(z)$ is a digamma function, λ the dimensionless constant of the electron-phonon interaction, $A_{\parallel} = (A_x, A_y)$, A_z is the vector potential, d the distance between layers (12 \AA in $\text{TaS}_2(\text{Py})_{1/2}$), D_{\parallel} and D_{\perp} are the mobilities along and perpendicular to the layers, and the z axis is directed perpendicular to the layers. If the condition $\hbar D_{\perp}/d^2 \ll T - T_c$ is satisfied for $T > T_c$, or the condition $\hbar D_{\perp}/d^2 T_c \ll (H - H_{c2}(0))/H_{c2}(0)$ for $H > H_{c2}(0)$ and $T = 0$, then transitions of electrons between the layers can be neglected and the fluctuations of the diamagnetic moment become two-dimensional. In $\text{TaS}_2(\text{Py})_{1/2}$, the critical temperature $T_c \approx 3.5^\circ \text{K}$ and $\hbar D_{\perp}/d^2 < 0.017^\circ \text{K}$ according to the estimates of^[4], so that the fluctuations lose their two-dimensional character only in the very immediate vicinity of the transition

point. Below we shall consider only the region of two-dimensional fluctuations, and omit the term in (3) that is proportional to D_{\perp} . Then we obtain for the free energy density from (1), (2), and (3)

$$\mathcal{F} = \frac{2eH_{\perp}}{\pi^2 \hbar c d} \sum_{n=0}^{\infty} \int_0^{\infty} d\omega \left[\frac{1}{2} + \frac{1}{e^{\beta \hbar \omega} - 1} \right] \text{Im} \ln \mathcal{D}(\omega, n), \quad (4)$$

$$\mathcal{E}_{\pm}(\omega, n) = i\omega \pm 2i\mu H + 4eD_{\parallel} H_{\perp} \left(n + \frac{1}{2} \right) / c. \quad (5)$$

According to (1), (4), the free energy represents the sum of the free energy \mathcal{F}_T associated with the thermal fluctuations (the terms $(e^{\beta \omega} - 1)^{-1}$ in the square brackets) and the energy \mathcal{F}_0 connected with the zero-point oscillations (the term $1/2$ in the square brackets). Inasmuch as the two depend quite differently on temperature and field, we shall consider them separately.

3. THERMAL FLUCTUATIONS IN A TWO-DIMENSIONAL SYSTEM

From the expression for the free-energy density of thermal fluctuations

$$\mathcal{F}_T = \frac{2eH_{\perp}}{\pi^2 \hbar c d} \sum_{n=0}^{\infty} \int_0^{\infty} d\omega \frac{\text{Im} \ln \mathcal{D}(\omega, n)}{e^{\beta \hbar \omega} - 1} \quad (6)$$

it is seen that the basic contribution to the integral over ω is made by frequencies $\omega \lesssim T$, and that in the expression for $\mathcal{D}(\omega, n)$ we need leave only the term linear in $\omega/4\pi T$, using the small numerical parameter $1/2\pi$. In the analysis of the thermal fluctuations, we limit ourselves to the case of fields that are perpendicular to the layers. Then the paramagnetic effect can be neglected in (5). We replace the summation over n in (6) by an Euler-Maclaurin expansion and carry out the integration over ω . We can then represent the part of the free-energy density that depends on the field in the form

$$\mathcal{F}_T = - \frac{T^2}{4\pi^2 \hbar D_{\parallel} d} \left\{ 4h^2 \sum_{m=0}^{\infty} \tilde{B}_m (2h)^{2m} \frac{d^{2m}}{dh^{2m}} g'(h) Y[g(h)] - \int_0^h x g'(x) Y[g(x)] dx \right\}, \quad h = \frac{\Delta_0 H_{\perp}}{TH_{c2\perp}(0)}, \quad H_{c2\perp}(0) = \frac{c\Delta_0}{2eD_{\parallel}}, \quad (7)$$

$$\tilde{B}_m = B_{2m+2} / (2m+2)!, \quad Y(g) = \ln(g/2\pi) - \pi/g - \psi(g/2\pi),$$

$$g(x) = 4\pi \left[\psi(1/2 + x/4\pi) - \psi(1/2) + \ln(T/T_c) \right] / \psi'(1/2 + x/4\pi),$$

where B_n are the Bernoulli numbers and $H_{c2\perp}(0)$ is the upper critical field perpendicular to the layers at $T = 0$. For the diamagnetic moment M_T , we have from (7)

$$M_T = - \frac{eT}{2\pi^2 \hbar c d} \left[h - \frac{d}{dh} \sum_{m=0}^{\infty} \tilde{B}_m (2h)^{2m+2} \frac{d^{2m}}{dh^{2m}} g'(h) Y[g(h)] \right]. \quad (8)$$

At very weak fields $h \ll \ln(T/T_c)$, it suffices to consider in (8) only the first term in the sum over m . We then obtain for the temperature dependence of the diamagnetic susceptibility χ_T

$$\chi_T = - \frac{e^2 D_{\parallel}}{3\pi^2 \hbar c^2 d} \left[\ln \left(\frac{4}{\pi^2} \ln \frac{T}{T_c} \right) - \frac{\pi^2}{8 \ln(T/T_c)} - \psi \left(\frac{4}{\pi^2} \ln \frac{T}{T_c} \right) \right]. \quad (9)$$

For $T - T_c \ll T_c$, we have from (9)

$$\chi_T = - \frac{e^2 D_{\parallel}}{24 \hbar c^2 d} \frac{T_c}{T - T_c} \quad (10)$$

in correspondence with the result of Schmid^[7] for a two-dimensional system.

For $h/2\pi \ll 1$, we can represent the total sum over m in the expression for M_T in the form of an integral, since in this case $g(h)$ has the simple form

$$g(h) = h + \frac{8}{\pi} \ln \frac{T}{T_c}. \quad (11)$$

Such an approximation for $g(h)$ is equivalent to expanding the digamma function ψ in the expression for \mathcal{D} in a series in $1/T$ and discarding all terms except the linear one. Maki and Takayama^[6] used this approximation with arbitrary h for the three-dimensional case.^[6] It is seen from (8) that it is valid only for not very strong fields, $h/2\pi \ll 1$. It is shown in the Appendix that expression (8), with the function $g(h)$ determined in (11), can be transformed to

$$M_T = - \frac{eT}{24\pi^2 \hbar c d} \left[h Y \left(h + \frac{8}{\pi} \ln \frac{T}{T_c} \right) - \pi \frac{d}{dh} h \int_0^{\infty} \frac{dx}{x} \exp \left[-x \left(h + \frac{8}{\pi} \ln \frac{T}{T_c} \right) \right] \right] \times \left(\text{cth} \pi x - \frac{1}{\pi x} \right) \left(\text{cth} h x - \frac{1}{hx} \right) \quad (12)$$

For $h/4\pi \gg 1$, we have $g(h) \approx 4\pi h \ln h$ and from (8)

$$M_T = - \frac{eT}{24\pi \hbar c d} \frac{1}{h \ln h} \left[1 + o \left(\frac{1}{\ln h} \right) \right]. \quad (13)$$

It is seen from (13) that the diamagnetic moment associated with the thermal fluctuations vanishes when the magnetic field H begins to exceed a value of the order of $H_{c2\perp}(0)$.

4. ZERO POINT OSCILLATIONS OF COOPER PAIRS IN A TWO-DIMENSIONAL SYSTEM

We have for the free-energy density of the zero-point oscillations of the Cooper pairs

$$\mathcal{F}_0 = \frac{eH_{\perp}}{\pi^2 \hbar c d} \sum_{n=0}^{\infty} \int_0^{\infty} d\omega \text{Im} f \left[-i\omega + 2hT \left(n + \frac{1}{2} \right) \right], \quad (14)$$

$$f[-i\omega + 2hT(n+1/2)] = \ln \mathcal{D}^{-1}(-\omega, n).$$

For summation of the function $f(\omega, n)$ over n , we use the Euler-Maclaurin formula in the form

$$\sum_{n=0}^{\infty} f(n) = \int_0^{\infty} f(x) dx + \frac{1}{2} f(0) + i \int_0^{\infty} \frac{f(ix) - f(-ix)}{e^{2\pi x} - 1} dx. \quad (15)$$

As above, in the use of the Euler-Maclaurin formula, we assume that the actual function $f(\omega, x)$ and all its derivatives tend to zero as $x \rightarrow \infty$ and $\omega \rightarrow \infty$, so that the first integral on the right side of (15) converges. The function (2) as used by us is approximate, and these conditions are not satisfied for it: $f(\omega, x)$ does not tend to zero as $x \rightarrow \infty$ or $\omega \rightarrow \infty$, and the first integral on the right side of (15) diverges. This is because the diffusion approximation used to obtain (2) is invalid at high frequencies $\omega \gtrsim \hbar/\tau$ and large momenta (larger n); however, we are interested only in that part of the free energy which depends on the field. It is determined by the values of $f(\omega, x)$ at finite x . In the region $2hT_x \lesssim \hbar/\tau$, the function (2) correctly reproduces the behavior of the actual function $f(\omega, x)$ and we can use it for calculation of the diamagnetic properties. Separating from the first integral in (15) that part which depends on the field, and keeping it only, we obtain

$$\mathcal{F}_0 = - \frac{hT}{4\pi^2 \hbar D_{\parallel} d} \text{Im} \int_0^{\infty} d\omega \left\{ \int_0^i dx [f(hTx - i\omega) - f(hT - i\omega)] - \frac{1}{2\pi} \int_0^{\infty} dx [f(2ihTx + hT - i\omega) - f(-2ihTx + hT - i\omega)] \frac{d}{dx} \ln(1 - e^{-2\pi x}) \right\}, \quad (16)$$

$$f(x) = \ln \left\{ \lambda \left[\psi \left(\frac{1}{2} + \frac{x}{4\pi T} \right) - \psi \left(\frac{1}{2} \right) + \ln \frac{T}{T_c} \right] \right\}.$$

Integrating by parts in the integrals over x , we have the possibility of carrying out the integration over the frequency ω . However, it was noted above that the function $f(\omega)$ diverges as $\omega \rightarrow \infty$ if we use expression (2) for \mathcal{D} . As in [3], we take the quantity \hbar/τ as the upper limit of integration over ω , assuming that the actual function $f(\omega)$ falls rapidly to zero at higher frequencies. Then (16) transforms to

$$\mathcal{F}_0 = -\frac{\hbar^2 T^2}{4\pi^2 \hbar D_{\parallel} d} \operatorname{Re} \left\{ \int_0^{\hbar/\tau} dx dx [f(\hbar T x - i\hbar \tau^{-1}) - f(x)] \right. \\ \left. + \frac{1}{\pi} \int_0^{\hbar/\tau} dx \ln(1 - e^{-\pi x}) [f(i\hbar T x + \hbar T - i\hbar \tau^{-1}) + f(-i\hbar T x + \hbar T - i\hbar \tau^{-1}) \right. \\ \left. - f(i\hbar T x + \hbar T) - f(-i\hbar T x + \hbar T)] \right\}. \quad (17)$$

It is seen from (17) that in the case of fields that are perpendicular to the layers, the contribution of the zero-point oscillations to the diamagnetic moment vanishes only when $\hbar T \gg \hbar/\tau$, i.e., when the field H exceeds the value $H_S = \Phi_0/l^2$.

For perpendicular fields $H \ll H_S$, we get a very slow decrease in the diamagnetic moment with increasing H from (17):

$$M_0 = -\frac{e\hbar T}{6\pi^2 \hbar c d} \left[\operatorname{Re} \ln \frac{Z(T)}{\ln(T/T_c) + \pi\hbar/8} - \left(1 + \frac{8}{\pi\hbar} \ln \frac{T}{T_c}\right)^{-1} \right] \quad (18)$$

for $\hbar/2\pi \ll 1$ and

$$M_0 = -\frac{e\hbar T}{6\pi^2 \hbar c d} \left[\operatorname{Re} \ln \frac{Z(T)}{\ln(\hbar T/\Delta_0)} - \frac{1}{\ln(\hbar T/\Delta_0)} \right] \quad (19)$$

for $\hbar/2\pi \gg 1$. Here

$$Z(T) = \psi \left(\frac{1}{2} + \frac{i\xi_0 \Delta_0}{4lT} \right) - \psi \left(\frac{1}{2} \right) + \ln \frac{T}{T_c}.$$

It follows from (18) that in fields for which $\pi\hbar \ll 8 \ln(T/T_c)$, the diamagnetic susceptibility χ_0 is determined by the expression

$$\chi_0 = -\frac{e^2 D_{\parallel}}{3\pi^2 \hbar c^2 d} \operatorname{Re} \ln \frac{Z(T)}{\ln(T/T_c)}. \quad (20)$$

For temperatures close to T_c , the contribution of the thermal fluctuations (9) to the total fluctuation diamagnetic susceptibility χ far exceeds the contribution of the zero-point oscillations (20). However, far from the critical temperature, the situation changes and at $T = 10 T_c$ for $\xi_0/l = 11.4$ we have $\chi = \chi_T + \chi_0 = -0.554 e^2 D_{\parallel} / 3\pi^2 \hbar c^2 d$ and $\chi_0 = 5.6 \chi_T$, and for $\xi_0/l = 5.7$ we get $\chi = -0.314 e^2 D_{\parallel} / 3\pi^2 \hbar c^2 d$ and $\chi_0 = +2.7 \chi_T$.

We now compare the contribution of the zero-point oscillations of the Cooper pairs to the diamagnetic susceptibility with the Landau diamagnetism of normal electrons. For fields with $\omega_H = eH_{\perp}/mc \gg D_{\perp}/d^2$ this condition is satisfied in $\text{TaS}_2(\text{Py})_{1/2}$ at least for $H \gg 1$ kOe, the motion of the normal electrons in a magnetic field can be assumed to be two-dimensional, and we obtain for the Landau diamagnetic susceptibility¹⁾

$$\chi_L = -e^2/12\pi mc^2 d. \quad (21)$$

It is seen from comparison of χ_0 with χ_L that for temperatures $T - T_c \sim T_c$ the contribution of the zero-point oscillations differs from the contribution of the normal electrons by a factor $(k_F l) \gg 1$. In the three-dimensional case, according to the estimate of Maki,^[3] the contribution of the zero-point oscillations is the same in order of magnitude as the diamagnetism of the normal electrons.

We now consider the fluctuations of the diamagnetic moment at the temperature $T = 0$ in fields $H \gtrsim H_{c2\perp}(0)$. In this case, the fluctuation diamagnetic moment is de-

termined only by the zero-point oscillations, since the contribution of the thermal fluctuations at $T = 0$ falls off in proportion to $(T/T_c)^2$.

For perpendicular fields $H \ll H_S$, the function $f(x) = \ln[\lambda \ln(x/\Delta_0)]$, and we get from (17)

$$M_0(H) = -\frac{e^2 D_{\parallel} H}{\pi^2 \hbar c d} \left[\ln \frac{[\ln(\hbar/\tau \Delta_0)]^{\hbar}}{\eta} \right. \\ \left. - \frac{2}{\pi} \int_0^{\hbar/\tau} dx \ln(1 - e^{-\pi x}) \frac{\eta^{+1/2} \ln(1+x^2)}{[\eta^{+1/2} \ln(1+x^2)]^2 + \operatorname{arctg}^2 x} \right], \quad (22)$$

where $\eta = \ln[H/H_{c2\perp}(0)]$. Upon approach of H to $H_{c2\perp}(0)$ in the region of two-dimensional fluctuations, the moment M_0 increases logarithmically:

$$M_0(H) = -\frac{e^2 D_{\parallel} H}{\pi^2 \hbar c^2 d} \ln \frac{[\ln(\hbar/\tau \Delta_0)]^{\hbar} H_{c2\perp}^2(0)}{\pi [H - H_{c2\perp}(0)]^2}. \quad (23)$$

In the region of three-dimensional fluctuations ($\eta \ll \hbar D_{\perp}/d^2 T_c$), the logarithmic increase ceases and the diamagnetic moment tends to a finite value as $H \rightarrow H_{c2\perp}(0)$. For $H_{c2\perp}(0) \ll H \ll H_S$, we get from (29):

$$M_0(H) = -\frac{e^2 D_{\parallel} H}{\pi^2 \hbar c^2 d} \left[\ln \frac{[\ln(\hbar/\tau \Delta_0)]^{\hbar}}{\eta} + \frac{1}{3\eta} \right]. \quad (24)$$

Thus, the moment M_0 falls very slowly with increasing H and disappears in fields exceeding H_S . The strong dependence of the fluctuation moment M_0 on H near $H_{c2\perp}(0)$ allows us to separate it from all the remaining contributions to the magnetic moment (the contribution of the normal electrons is proportional to H and the contribution of paramagnetic impurities does not depend on H for $\mu H \gg T$).

It is also of interest to consider the fluctuations of the diamagnetic moment at $T = 0$ in fields directed nearly parallel to the layers, when $H_{\perp} \ll H_{c2\perp}(0)$. The superconducting state is destroyed in this case by the paramagnetic effect, and a transition of the first kind to the normal state takes place in a field $H_p = \Delta_0/\mu\sqrt{2}$. The fluctuations of the diamagnetic moment above the field $H_0 = H_p/\sqrt{2}$ (in the range from H_0 to H_p the normal state is metastable) are determined by the expression

$$M_0 = -\frac{e^2 D_{\parallel} H_{\perp}}{3\pi^2 \hbar c^2 d} \ln \frac{\ln(\hbar/\tau \Delta_0)}{\ln(H/H_0)} \quad (25)$$

for fields $H \ll \pi \xi_0 H_0/l$, and the diamagnetic moment vanishes only in fields exceeding $H_p \xi_0/l$.

5. DISCUSSION OF THE EXPERIMENTAL DATA FOR FLUCTUATIONS IN $\text{TaS}_2(\text{Py})_{1/2}$

According to the results obtained above, the total fluctuation diamagnetic susceptibility of dirty layer superconductors with Josephson tunneling between the layers above T_c is inversely proportional to $T - T_c$ at $\hbar D_{\perp}/d^2 \ll T - T_c \ll T_c$, and it then falls off slowly with increase in the temperature. The diamagnetic fluctuation moment is strongly dependent on the value of the field for $T - T_c \lesssim T_c$ since the thermal fluctuations make the chief contribution in this range of temperatures. At higher temperatures, the diamagnetic moment falls off much more slowly with increasing field, since in this region the contribution of the zero-point oscillations of the Cooper pairs is predominant and that depends weakly on the value of the field H at $H \ll H_S$. Just such a qualitative picture of the diamagnetic fluctuations above T_c has been observed experimentally in $\text{TaS}_2(\text{Py})_{1/2}$ according to the communication of DiSalvo, Geballe, Menth and Gamble.^[8,9] Near T_c , Prober, Beasley and

Schwall^[10] obtained the dependence $\chi \sim 1/(T - T_c)$ experimentally; however, the coefficient of proportionality turned out to be less than that predicted theoretically by a factor of about 6–10. We note that the structural transition at 80°K, which is accompanied by a peak in the magnetic susceptibility, precludes quantitative estimation of the contribution of the diamagnetic fluctuations above T_c in the intercalated compounds of TaS₂.^[8] This difficulty is removed in measurements of diamagnetic fluctuations in fields $H \gtrsim H_{c2\perp}(0)$ and at temperatures $T \ll T_c$. As has already been noted above, such measurements would have given direct information on the zero-point oscillations of the Cooper pairs in the normal state of the superconductor.

Evidently the zero-point oscillations also give an important contribution to the fluctuation conductivity above the transition point. The very slow growth in resistance of TaS₂(Py)_{1/2} upon an increase in the magnetic field above H_{c2} for field directions that are close to parallel can be connected with this effect.^[5]

6. DIAMAGNETIC FLUCTUATIONS IN SMALL PARTICLES

The diamagnetic fluctuations in small superconducting particles with radii R small in comparison with the correlation length have been calculated with the help of the generalized static approximation of Ginzburg-Landau.^[11,12] However, the static approximation becomes inadequate far from T_c . Account of the dynamics of the fluctuations far from T_c leads to rapid decay of the thermal fluctuations with increasing $(T - T_c)$. This decay, however, is compensated by the contribution of the zero-point oscillations, thanks to which the diamagnetic fluctuations in the small particles fall off as slowly as in quasi two-dimensional systems.

We shall take the dynamical corrections to the diamagnetic fluctuations into account only in the region in which these corrections are large, i.e., outside the critical region (for temperatures $T - T_c \gg (T_c/N(0)\Omega)^{1/2}$, where $N(0)$ is the density of the electron states and Ω the volume of the particle). Outside the critical region, the fluctuations are small, and in the calculation of the free energy we can limit ourselves to terms of lowest order only in the amplitude of the fluctuating field. Just this approximation was used above in the present paper. For the propagation function of the fluctuating field \mathcal{D} in Eq. (1), we have, in the case of small particles,

$$\mathcal{D}(\omega, \mathbf{A}) = \lambda^{-1} \left\{ \psi \left[\frac{1}{2} + \frac{i\omega - \hbar D (\nabla - 2ie\mathbf{A}/\hbar c)^2}{4\pi T} \right] - \psi \left(\frac{1}{2} \right) + \ln \frac{T}{T_c} \right\}^{-1} \quad (26)$$

and in the calculation of the free energy we can neglect the space derivatives in Eq. (26). Then the calculation of the trace in (1) reduces to an averaging of the quantity $\ln \mathcal{D}(\omega, \mathbf{A})$ over the spatial coordinates. In calculating the contribution of the thermal fluctuations, it suffices to take into account only the linear term in the expansion of the digamma function in (26) in the series in $1/T$. Then the susceptibilities χ_T and χ_0 have the same temperature dependences as in layer superconductors (see (9) and (20)):

$$\chi_T = -\frac{8De^2R^2}{5\pi\hbar c^2\Omega} \left[\ln \left(\frac{4}{\pi^2} \ln \frac{T}{T_c} \right) - \frac{\pi^2}{8 \ln(T/T_c)} - \psi \left(\frac{4}{\pi^2} \ln \frac{T}{T_c} \right) \right],$$

$$\chi_0 = -\frac{8De^2R^2}{5\pi\hbar c^2\Omega} \operatorname{Re} \ln \frac{\psi(1/2 + i\hbar/4\pi T\tau) - \psi(1/2) + \ln(T/T_c)}{\ln(T/T_c)}, \quad (27)$$

$$\chi = \chi_T + \chi_0.$$

At $T = 0$, due to the zero-point oscillations, the diamagnetic moment is different from zero for fields that are greater than the critical field $H_{c2}^*(0) \approx \Phi_0/R\sqrt{l\xi_0}$. At $H \gtrsim H_{c2}^*(0)$, the moment amounts to about $De^2R^2\hbar/\hbar c^2\Omega$ and vanishes only when the value of the field begins to exceed Φ_0/lR .

In conclusion, the author expresses his gratitude to V. L. Ginzburg and participants in his seminar for a useful discussion of the work.

APPENDIX

We can transform the sum over m in the right side of (8) with $g = h + \xi$ in the following fashion:

$$\begin{aligned} & 4h^2 \sum_{m=0}^{\infty} B_m(2h)^{2m} \frac{d^{2m}}{dh^{2m}} \left[\ln \frac{h+\xi}{2\pi} - \frac{\pi}{h+\xi} - \psi \left(\frac{h+\xi}{2\pi} \right) \right] \\ &= 8h^2 \sum_{m=0}^{\infty} B_m(2h)^{2m} \frac{d^{2m}}{dh^{2m}} \int_0^{\infty} \frac{y dy}{(e^y-1)[y^2+(h+\xi)^2]} \\ &= 8h^2 \sum_{m=0}^{\infty} B_m(2h)^{2m} \frac{d^{2m}}{dh^{2m}} \int_0^{\infty} \frac{dy}{e^y-1} \int_0^{\infty} dx e^{-x(h+\xi)} \sin xy \\ &= \pi h \int_0^{\infty} \frac{dx}{x} e^{-x(h+\xi)} \left(\operatorname{th} \pi x - \frac{1}{\pi x} \right) \left(\operatorname{th} hx - \frac{1}{hx} \right). \end{aligned}$$

Note added in proof (April 22, 1973): In a recent paper, R. A. Klemm, M. R. Beasley, and A. Luther, Phys. Rev. 8B, 5072 (1973) also calculated the fluctuations of the diamagnetic moment in dirty layer superconductors above the temperature T_c , and the results of the calculations for the purely two-dimensional case agree with those obtained in the present work. However, Klemm, Beasley and Luther assume that the contribution of the zero-point oscillations to the diamagnetic susceptibility (20) is practically independent of the temperature and therefore cannot be observed experimentally. Actually, because of the zero-point oscillations, χ changes upon an increase in the temperature from $2T_c$ to $10T_c$ by an amount $\approx e^2 D_{\parallel} / 3\pi^2 \hbar c^2 d$, which is noticeable at the accuracy of measurement achieved in [9,10]. For TaS₂(Py)_{1/2} at $2T_c$ to $20T_c$, $\xi_0/l = 5.7$, and $v_F = 1.1$ cm/sec [4], we obtain, with account of the zero-point oscillations, a slow decrease of χ with increase in the temperature: $\chi_g \approx 0.7 \times 10^6 / (T - T_c)$. This dependence is close to that which was experimentally in [9].

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