Effect of spin fluctuations on the superconducting transition in UPt_3

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The role played by spin fluctuations in forming the superconducting state in heavy-fermion compounds is discussed. A mechanism involving these fluctuations is offered as a possible explanation of the splitting of the phase transition in UPt_3. It is suggested that anisotropic singlet superconductivity occurs in UPt_3 as the result of spin correlations at the Fermi surface near an antiferromagnetic instability. If sufficiently large, the spin fluctuations lead to splitting of the superconducting transition. Discrepancies between the weak-link theory and experimental results on the jumps in the specific heat are discussed. The effect of the superconducting state on the antiferromagnetism in UPt_3 is also discussed.

1. INTRODUCTION

The origin of the superconductivity in UPt_3 and other heavy-fermion compounds has been under discussion for a fairly long time now, but has yet to be finally resolved. The most popular interpretation these days is that singlet anisotropic superconductivity (i.e., the spin of the Cooper pair is $S=0$; this is a d pairing) operates in these structures. Although that interpretation has numerous pieces of experimental support, alternative explanations are not completely ruled out. It has been shown[1–3] that spin correlations near an antiferromagnetic instability can lead to a d pairing. Experiments on neutron scattering in UPt_3 do indeed indicate such correlations.[4] An antiferromagnetic transition at $T_N = 5$ K has also been observed. The magnetic moments corresponding to antiferromagnetic order are exceedingly small [$\mu = (0.01–0.02)\mu_B$]. Measurements reveal no jump in the specific heat in the course of this transition. As the temperature is lowered, the magnetic moments which arise at T_N increase down to $T_c^+ = 0.5$ K, which is the temperature of the first superconducting transition. Below this point, the moments stop increasing. At $T_c^- = 0.45$ K, a second superconducting transition occurs in UPt_3, according to measurements of the jumps in the specific heat, the ultrasound velocity, etc. Many theoretical papers have attributed the splitting of the superconducting transition to interaction between the superconducting order parameter and the antiferromagnetic order parameter. In these theories the superconductivity is usually described by the Ginzburg–Landau functional

$$F = \int dV[-\alpha\tau|\psi|^2 + \beta_1|\psi|^4 + \beta_2|\psi^2|^2 + F_{int}]. \quad (1)$$

where the complex vector $\psi = (\psi_1,\psi_2)$ is the order parameter of the superconductor, and

$$F_{int} = \gamma|\psi \mathbf{M}|^2, \quad (2)$$

where $\mathbf{M} = (M_1,M_2)$, is the antiferromagnetic order parameter.

It is not difficult to see that the interaction F_{int} causes local disruption of the perfect crystal symmetry ($D_{6h} \to D_{2h}$ in the case of UPt_3). If we assume a spatially uniform distribution of the vector $\mathbf{M}$, the solutions for ψ are also uniform. In a "pure" superconductor (i.e., one with $|\mathbf{M}| = 0$), the solution for ψ is $\psi = (1,i)$, and the perfect crystal symmetry is not disrupted. However, the appearance of a nonzero vector $\mathbf{M}$ has the consequence that the superconducting transition at $T = T_{c1}$ goes to the state $\psi = (1,0)$ (phase 1), while a second transition occurs at $T = T_{c2}$ to the state $\psi = (1,i\varepsilon)$; $\varepsilon < 1$ (phase 2; see Ref. 5, for example). Phases 1 and 2 are asymmetric under rotations in the basal plane of the hexagonal crystal. In the case of UPt_3, this circumstance means in particular that the upper critical field H_{c2} is anisotropic in the basal plane, but this conclusion contradicts experiment. If we instead consider a nonuniform distribution of the vector $\mathbf{M}$, we do not run into this contradiction, but the effective interaction is weaker in this case[5,6]—too weak to explain the observed splitting $T_c^+ - T_c^-/T_c^+ = 0.1$.

In taking this approach, one is essentially assuming that the distribution of magnetic moments is already frozen at the superconducting transition temperature and that fluctuations can be ignored. It follows from this assumption, in particular, that the superconductivity has only a weak effect on the antiferromagnetism. This conclusion contradicts neutron-scattering experiments.[4] These experiments indicate that the antiferromagnetic order parameter ceases to increase upon the transition to the superconducting state, so the superconductivity suppresses the antiferromagnetism.

There is still some uncertainty regarding the actual width of the band of electrons involved in forming the antiferromagnetic state, but the density of states at the Fermi level corresponds to a width $E_F = 10–20$ K. The weak-link theory seems to be insufficient to describe the antiferromagnetism in UPt_3, since the quantity T_N/E_F is not small. The width of the fluctuation region of the phase transition in the quasi-2D case (which is apparently the case which holds in UPt_3) is

$$\tau = (T_N - T)/T_N \approx (T_N/E_F)|\ln(t)|. \quad (3)$$

Here $t \approx J/T_c$, where J is the jumping integral between planes,[7] and $|\ln(t)|$ can be numerically large enough to make the quantity τ of order unity. Fluctuations must therefore be taken into account all the way to absolute zero.

The mechanism proposed below for the splitting of the superconducting transition in UPt_3 is based on a consideration of spin fluctuations in a calculation of the free energy.

Brinkman *et al.*[8] have shown that spin fluctuations in

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He^3, provided that they are sufficiently large, make the AM phase preferable to the BW phase from the energy standpoint. In contrast with ^3He, in which the superfluidity is of the p type, it is believed that UPt_3 is a d-type superconductor, but there are formal analogies in the descriptions of these two substances. A parameter of the theory is the quantity $\alpha = T_c/E_F \cdot (1-\overline{I})$, where $\overline{I}$ is the average interaction constant, which will be determined below. For both substances the quantity $1-\overline{I}$ is small, and the parameter α, which is equal to the ratio of the fluctuation component of the free energy to the ordinary component, may be of order unity.

We will calculate this component to terms of fourth order in ψ. The contribution of fluctuations with a wave vector $q = 2k_F$ gives us a ratio of coefficients β_2/β_1 which is the same as in the weak-link theory ($\beta_2/\beta_1 = 1/2$), but this ratio changes under the condition $q < 2k_F$. Furthermore, the overall coefficient β_2 may change sign, causing a splitting of the transition in the superconductor. In the following section of this paper we calculate the absolute value of the fluctuation component. In Sec. 3 we show that the fluctuation component alters the symmetry of the solution for the superconducting order parameter, and we discuss possible scenarios for the onset of superconductivity in UPt_3. We conclude with a discussion of the effect of superconductivity on the spectrum of paramagnons in UPt_3, in particular, suppression of the antiferromagnetic order parameter by the superconducting order parameter.

2. CALCULATION OF THE FREE ENERGY; THE FLUCTUATION CONTRIBUTION

We assume that d-type superconductivity, due to the exchange of paramagnons, operates in UPt_3.

It was shown that in Refs. 1–3 that depairing is preferable to s or p pairing if the interaction $I\chi_0(q)$ is of the form

$$I\chi_0(q) = J_0 - J_1\gamma_q, \quad \gamma_q = 2[\cos(q_x a) + \cos(q_y a) + \cos(q_z a)], \qquad (4)$$

where J_0 and J_1 are constant such that $\chi_0(q)$ has a minimum at $q \neq 0$ ($q \sim 2k_F$). A p-type superfluid state occurs in ^3He, and the interaction has a maximum at the wave vector $q = 0$. This circumstance has important implications for the symmetry of the solution. The calculation of the absolute value of the fluctuation contribution in the case of d pairing is nearly the same as in the case of ^3He.

Following Ref. 8, we consider the standard expression for the spin-fluctuation component of the free-energy in the random phase approximation:

$$\Delta F^{SF} = -T\sum_{\mathbf{k},\omega_n}\left[\frac{I\delta\chi(\mathbf{k},\omega_n)}{1 - I\chi(\mathbf{k},\omega_n)}\right]^2, \qquad (5)$$

where

$$\chi_{\alpha\beta}(\mathbf{q}, i\omega_n) = 2\int_0^\infty \exp(i\omega_n\tau)\langle T_\tau[\hat{M}_\alpha(\mathbf{q},\tau)\hat{M}_\beta(-\mathbf{q},0)]\rangle d\tau$$

is the susceptibility, $\hat{M}$ is the magnetic moment operator, and $\delta\chi = \chi^S - \chi^N$ is the difference between the susceptibilities in the superconducting and normal states. To calculate the absolute value of the change in the free energy, we assume that the order parameter Δ is a scalar; we can then write

$$\Delta F^{SF} \approx \frac{N(0)\Delta^4}{\pi T E_F(1-\overline{I})}. \qquad (6)$$

Here $\overline{I} = \max I\chi_0(q)$. We of course cannot calculate the numerical coefficient in (4); in order to do that we would need to know the details of the interaction χ, and we would also need to know the exact solution $\Delta(\mathbf{k}, i\omega_n)$ in weak-link theory. The ordinary component of the free energy is $\Delta F^{(4)} = N(0)\zeta(3)\Delta^4/(\pi T)^2$, we then have

$$-\frac{\Delta F^{SF}}{\Delta F^{(4)}} = \frac{T}{E_F(1-\overline{I})} = \alpha\frac{T}{T_c}. \qquad (7)$$

If $\alpha \sim 1$ and $T \sim T_c$, the fluctuation component is comparable to the ordinary component. As $T \to 0$, the fluctuation component becomes negligible. A pronounced increase in the spin-fluctuation component occurs near the antiferromagnetic transition. In the random phase approximation, the effective interaction is

$$I_{eff}(\mathbf{q},\omega) = \frac{I}{1 - I\chi(\mathbf{q},\omega)}. \qquad (8)$$

Right at the transition point we have $1 - I\chi_0(\mathbf{Q},0) = 0$, where $\mathbf{Q}$ is the antiferromagnetic vector, and the fluctuations are large. When the antiferromagnetic parameter acquires a finite value, the denominator in (6) becomes finite, although small (the magnetic moments are $\mu \approx 0.01\mu_B$). Because of this small value, fluctuations must be taken into account. In the following section of this paper we consider the contribution of spin fluctuations for various wave vectors $\mathbf{q}$, and we calculate the ratio of Ginzburg–Landau coefficients β_2/β_1. This ratio determines the symmetry of the solution.

3. EFFECT OF SPIN FLUCTUATIONS OF THE SYMMETRY OF THE SOLUTION; POSSIBILITY OF SPLITTING OF THE PHASE TRANSITION

We first consider the symmetry of the ordinary superconducting component of the free energy:

$$\Delta F^{(4)} = T\sum_{\omega_n}\int\frac{(\Delta^+\Delta)^2}{(\omega_n^2 + \xi_{\mathbf{k}}^2)^2}d^3k. \qquad (9)$$

Following Millis *et al.*,[9] we separate the frequency part from the momentum part in the expression for Δ:

$$\Delta_{\alpha\beta}(\mathbf{k}, i\omega_n) = i\sigma^y_{\alpha\beta}\Delta(\omega_n)\eta(\mathbf{k}). \qquad (10)$$

We also expand the function η in the basis functions of the $2D$ representation of the crystal symmetry group of a hexagonal crystal:

$$\eta(\mathbf{k}) = \eta_1\phi_1(k) + \eta_2\phi_2(\mathbf{k}),$$
$$\phi_1(\mathbf{k}) = k_x k_z, \quad \phi_2(\mathbf{k}) = k_y k_z. \qquad (11)$$

We assume $\boldsymbol{\eta} = (\eta_1, \eta_2)$, then substituting (8) and (9) into (7), we find

$$\Delta F^{(4)} = \zeta(3)\frac{\Delta^4}{(\pi T)^2}(|\eta|^4 + \frac{1}{2}|\eta^2|^2). \quad (12)$$

We now wish to calculate the fluctuation component. To do this, we go back to expression (3). To calculate the contribution to the free energy of fourth order in Δ, we need to determine $\delta\chi(\mathbf{q},\omega_n)$ to within terms of second order in Δ. Using the standard expression for the susceptibility χ,

$$\chi(\mathbf{q}, \omega_m) = -\frac{T}{2}\sum_{\mathbf{p},n} [G(p_-)G(p_+) - F^+(p_+)F(p_-)],$$

$$p_\pm = (\mathbf{p} \pm q/2, \ \omega_n \pm \omega_m/2), \quad (13)$$

we find

$$\delta\chi(\mathbf{q}, \omega_m) = \int \Big\{ a(\mathbf{q}, i\omega_m)\Big[\eta\Big(\mathbf{p} + \frac{\mathbf{q}}{2}\Big)\eta^*\Big(\mathbf{p} + \frac{\mathbf{q}}{2}\Big) + \eta\Big(\mathbf{p} - \frac{\mathbf{q}}{2}\Big)\eta^*\Big(\mathbf{p} - \frac{\mathbf{q}}{2}\Big)\Big] + b(\mathbf{q}, i\omega_m)\eta\Big(\mathbf{p} - \frac{\mathbf{q}}{2}\Big)\eta^*\Big(\mathbf{p} + \frac{\mathbf{q}}{2}\Big)\Big\} d^3p. \quad (14)$$

The quantities a and b in (11) depend on the form of $\Delta(i\omega_n)$ and cannot be calculated for the general case. However, if Δ is independent of the frequency, then the quantities a and b become $D_2(m)$ and $D_1(m)$, respectively, to within a coefficient which is independent of ω_m (Ref. 8). Integrating in (11), we find an expansion of $\delta\chi$ in components of the vector η:

$$\delta\chi(\mathbf{q}, i\omega_m) = A(\mathbf{q}, i\omega_m)|\eta|^2 + B(\mathbf{q}, i\omega_m)(\eta\phi)(\eta^*\phi). \quad (15)$$

Without going through an explicit calculation of A and B, we can write

$$A(\mathbf{q}, i\omega_m) = 0, \quad |\mathbf{q}| = 2k_F.$$

The vectors $\mathbf{p} + \mathbf{q}/2$ and $\mathbf{p} - \mathbf{q}/2$ lie on the Fermi surface (they correspond to electron Green's functions), so if $|\mathbf{q}| = 2k_F$ holds then $\mathbf{p} = 0$. Taking the parity of $\eta(\mathbf{k})$ into account,[9] we find $\delta\chi(\mathbf{q}) \sim |\eta(\mathbf{q})|^2$; expression (14) then follows as a result. The ratio of coefficients $\beta_2/\beta_1 = 1/2$ has a value of 1/2 when spin fluctuations with a wave vector $q = 2k_F$ are taken into account. In other words, its value here is the same as in weak-link theory. We can now write a complete expression for ΔF^{SF}, derived under some rather severe limitations (χ and Δ are independent of the frequency, and all the electron momenta lie on the Fermi surface):

$$\Delta F^{SF} = -\frac{\Delta^4}{\pi T E_F(1 - \bar{I})}\int d\xi(1 - \xi^2)^2\chi^2(\xi)\{[P(1 + 3\xi^2) + Q(1 - 5\xi^2)]^2|\psi|^4 + \frac{1}{2}[P(1 - 5\xi^2) + Q(1 + 3\xi^2)]^2|\psi^2|^2\}. \quad (16)$$

Here $\xi = q/2k_F$, $P < Q$ (see the Appendix), and $\chi(\xi)$ is the susceptibility of the normal metal (which is now independent of the frequency). It follows in particular from (16) that we have $2\beta_2^{SF} > \beta_1^{SF}$ (β_2^{SF} and β_1^{SF} are the coefficients of the terms $|\eta^2|^2$ and $|\eta|^4$, respectively, in the expansion of the spin-fluctuation component of the free energy, ΔF^{SF}, in η). The complex expression for the free energy (the terms of fourth order) is

$$\Delta F^{(4)} = \beta_1(|\psi|^4 + \frac{1}{2}|\psi^2|^2) + \beta_1^{SF}|\psi|^4 + \beta_2^{SF}|\psi^2|^2. \quad (17)$$

Here $\beta_1 > 0$, $\beta_1^{SF} < 0$, and $\beta_2^{SF} < 0$. We introduce the parameters $\tilde{\alpha} = |\beta_1^{SF}/\beta_1|$ and $\varkappa = 2\beta_2^{SF}/\beta_1^{SF}$ ($\varkappa > 1$). We then have $\tilde{\alpha} \approx \alpha$. The parameter $\tilde{\alpha}$ falls off with decreasing temperature. Three scenarios for the onset of superconductivity are possible, depending on the parameters $\tilde{\alpha}$ and $\varkappa$ (Ref. 6).

1. There is a simple transition ($\tilde{\alpha} < 1/\varkappa < 1$). In this case we have $\beta_1^{\text{eff}} = \beta_1 + \beta_1^{SF} > 0$, $\beta_2^{\text{eff}} = \beta_1/2 + \beta_2^{SF} > 0$ at all temperatures below T_c. The transition is to a symmetric phase $\eta = (1,i)$ (the condition $|\eta^2| = 0$ is favored from the energy standpoint).

2. The transition splits ($1/\varkappa, \tilde{\alpha}(T_c) < 1$). In this case we have $\beta_1^{\text{eff}}(T_c) > 0$, $\beta_2^{\text{eff}}(T_c) < 0$, and the transition is to a real phase $\eta = (1,0)$ ($|\eta^2| = |\eta|^2$). At a sufficiently low temperature $T = T_{\text{cr}}$, the coefficient β_2^{eff} changes sign. This change in sign occurs because the fluctuation component is inversely proportional to the temperature, while the ordinary component is proportional to the square of the temperature [see (4) and (5)]. Let us single out the functional dependence $F_2(|\psi^2|)$, i.e., the part of the free energy which depends only on the invariant $|\psi^2|^2$. If the function F_2 has a minimum, then at a temperature T_c^- such that the conditions $T_c > T_c^- > T_{\text{cr}}$, and $F_2(|\psi(T_c^-)^2|) = \min F_2$ hold there will be a second superconducting transition: $\eta = (1,0) \to \eta = (1,i\varepsilon)$ (cf. Ref. 6). At the temperature T_{cr}, a third transition occurs: $\eta = (1,0) \to \eta = (1,i\varepsilon)$. There is the further possibility that this third transition does not occur, since the minimum in F_2 may survive the change in the sign of β_2^{eff} (this would be fully developed superconductivity, and the region of small values of Δ would no longer be important).

If F_2 is instead always a monotonic function then a first-order transition occurs at the temperature $T = T_{\text{cr}}$: $\eta = (1,0) \to \eta = (1,i)$.

3. There is a first-order transition ($\tilde{\alpha}(T_c) > 1$). In this case we have $\beta_1 < 0$, $\beta_2 < 0$. The functional is stable in this case, by virtue of the sixth-order coefficients. The superconducting transition is a first-order transition. This scenario may also apply to s-type superconductivity, but it is doubtful that it actually occurs. If it did, the implication would be that the fluctuation component would be greater than the ordinary component at small values of Δ.

4. CONCLUSION

We have examined the effect of spin fluctuations on the superconductivity in UPt_3. We have shown that fluctuations apparently must be taken into account in studies of heavy-fermion compounds, in which the width of the electron spectrum near the Fermi level is small. If the effect of the fluctuations is sufficiently large, the phase transition will split in a d-type superconductor. Within the framework of this model, of course, we cannot derive numerical expressions for the magnitude of the splitting of the transition, the jumps in the specific heat, etc. In order to do this, we would need to know accurate values of the interaction constants, the details of the spectrum, etc. At a qualitative level, however, it is possible to explain (for example) the relatively small splitting of the transition. The explanation is that the free energy is determined primarily by spin fluctuations with $q = 2k_F$ (in which case the effective interaction increases rapidly near the antiferromagnetic instability). It has been established that the relation $\beta_1 = 2\beta_2$ holds exactly for such excitations. If we were to analyze the contribution from such excitations alone, we would reach the conclusion that no splitting of any sort could occur: The superconductivity would arise in accordance with either the first or third scenario outlined above. Only by incorporating fluctuations with $q < 2k_F$ could we obtain splitting of the transition. However, the magnitude of the effective interaction, $I_{\rm eff}(q)$, falls off fairly rapidly at $q \neq 2k_F$, and the contribution of such excitations is not very significant, nor is the magnitude of the splitting. From this theory we draw the following conclusions, in particular: A finite superconducting gap leads to a suppression of the low-frequency part of the paramagnon spectrum. Incorporating spin fluctuations is equivalent to incorporating renormalization of the paramagnon spectrum in the transition to the superconducting state.

Millis *et al.*[9] have shown that excitations with frequencies $\omega < \omega_c \sim T_c \exp(J_0/J_1)$ suppress the superconductivity. A negative spin-fluctuation contribution to the free energy signifies a decrease in the density of states of low-frequency paramagnons. It is thus not difficult to draw the further conclusion that the superconductivity suppresses the antiferromagnetism: The suppression of the antiferromagnetic order parameter means that the isotropic susceptibility in the basal plane is joined by an anisotropic part at $\omega = 0$, and the latter is suppressed in the transition to the superconducting state.

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APPENDIX

Introducing the one-particle Green's functions

$$G(i\omega_n, p) = -\frac{i\omega_n + \xi_{\mathbf{p}}}{\omega_n^2 + \xi_{\mathbf{p}}^2 + |\Delta_{\mathbf{p}}|^2},$$

$$F(i\omega_n, p) = \frac{\Delta_{\mathbf{p}}}{\omega_n^2 + \xi_{\mathbf{p}}^2 + |\Delta_{\mathbf{p}}|^2},$$

substituting them into expression (13) for χ, and subtracting the normal-metal susceptibility, we find

$$\delta\chi(\mathbf{q}, i\omega_m) = \frac{N(0)}{4E_F T}\int \frac{d\phi}{2\pi}[|\Delta_{\mathbf{p}}|^2 A(\mathbf{q}, i\omega_m) + \Delta_{\mathbf{p}}\Delta_{\mathbf{p}'}B(\mathbf{q}, i\omega_m)].$$

The integration here is carried out under the conditions

$$\mathbf{p} - \mathbf{p}' = \mathbf{q}, \quad |\mathbf{p}| = |\mathbf{p}'| = p_F.$$

We can write explicit expressions for P and Q:

$$P = T\sum_n \int \frac{[i\omega_n + \xi(\mathbf{p})][i(\omega_n - \omega_m) + \xi(\mathbf{p}')]d^3p}{[\omega_n^2 + \xi(\mathbf{p})^2][(\omega_n - \omega_m)^2 + \xi(\mathbf{p}')^2]^2},$$

$$Q = T\sum_n \int \frac{d^3p\,d^3p'}{[\omega_n^2 + \xi(\mathbf{p})^2][(\omega_n - \omega_m)^2 + \xi(\mathbf{p}')^2]}.$$

The quantities P and Q are real and positive. Their specific values depend on the electron spectrum. In the simple case of a spherical Fermi surface, P and Q become the D_2 and D_1, respectively, of Ref. 8. The condition $D_2 < D_1$ holds. Now substituting the explicit expressions for Δ [see (10)] into the expression for $\delta\chi$, and integrating $|\Delta_{\mathbf{p}}|^2$ over the angle ϕ, we find

$$\langle|\Delta_{\mathbf{p}}|^2\rangle = \Delta^2(1 - \xi^2)[|\eta|^2(1 - \xi^2) + (\eta\nu)(\eta^*\nu)(5\xi^2 - 1)].$$

Correspondingly, we find

$$\langle\Delta_{\mathbf{p}}\Delta^*_{\mathbf{p}'}\rangle = \Delta^2(1 - \xi^2)[|\eta|^2(1 - \xi^2) - (\eta\nu)(\eta^*\nu)(3\xi^2 + 1)],$$

where $\xi = q/2k_F, \nu = \mathbf{q}/q$. We thus find

$$\delta\chi(\mathbf{q}, i\omega_m) = \Delta^2 \frac{N(0)}{4E_F T}(1 - \xi^2)\{|\eta|^2(1 - \xi^2)(P + Q) + (\eta\nu)(\eta^*\nu)[P(5\xi^2 - 1) - Q(3\xi^2 + 1)]\}.$$

Finally substituting the expression for $\delta\chi$ into (5), and integrating over the direction of the vector $\mathbf{q}$, we find the expression which we have been seeking [expression (16)].

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