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Spin fluctuations and superconductivity in UPt_3

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We attempt to assess the importance of spin fluctuations in the heavy-fermion system UPt_3, the most unambiguous evidence for which is the $T^3 \ln T$ term in the specific heat. We investigate whether other contributions, such as that from a peak in the electronic density of states or from the electron-phonon interaction, could account for the experimental data. We conclude that they cannot, although the data are consistent with the presence of both a $T^3 \ln T$ term and a density-of-states peak of width greater than about 60 K. We determine the input parameters for the paramagnon theory with a self-consistent method developed by Boring, Albers, Stewart, and Koelling for UAl_2 and we calculate the s- and p-wave pairing interactions. A one-band model favors p-wave pairing, while a two-band model leads to conventional s-wave superconductivity.

Since the discovery of its superconductivity[1] and the observation of a $T^3 \ln T$ temperature dependence in the specific heat,[1,2] the heavy-fermion system UPt_3 is considered to be the first case of the coexistence of spin fluctuations (SF) and superconductivity. It has also been suggested[1] that, in analogy to ^3He, the superconductivity might be of the "p" type. Here we examine the evidence for SF in UPt_3 and calculate the SF contribution to the pairing interaction to the extent possible in the absence of reliable band-structure input for the SF theory.

Although a number of observations,[3] for example, the T^2 temperature dependence of the susceptibility and resistivity, are consistent with the presence of SF, the least ambiguous seems to be the $T^3 \ln T$ term in the specific heat. While this term is in principle always present in a clean Fermi liquid,[4] it is much enhanced in a nearly ferromagnetic system. Such a term can also arise from the electron-phonon interaction; however, this contribution has the opposite sign from the SF term by virtue of the particular energy dependence of the electron self-energy.[5] Attributing the entire observed $T^3 \ln T$ term in UPt_3 to SF is thus actually an *underestimate* if the electron-phonon contribution is sizable. Also, the fact that this term disappears upon substitution of Pd for Pt (Ref. 6) is in accordance with the SF theory;[7] an electron-phonon contribution should be unaffected by impurity scattering.

de Visser, Franse, Menovsky, and Palstra[2] have fitted their specific-heat data in the temperature range 0–20 K with

$$C/T = \gamma + \beta^* T^2 + \delta T^2 \ln T \ , \tag{1}$$

where $\gamma = 422$ mJ (mol U)$^{-1}$K^{-2}, $\beta^* = -3.8$ mJ (mol U)$^{-1}$K^{-4}, and $\delta = 1.4$ mJ (mol U)$^{-1}$K^{-4}. Here $\beta^* = \beta - \delta \ln T^*$, where β is the coefficient of the lattice term and T^* is a characteristic temperature.

We first consider the question of whether the contribution of a peak in the density of states $N(E)$ per spin can lead to a specific-heat $C_{\text{band}}(T)$ that fits the data. We employ

$$C_{\text{band}}(T) = \left(\frac{2}{k_B T^2}\right) \int_{-\infty}^{\infty} N(E) E^2 \frac{\exp(E/k_B T)}{[1+\exp(E/k_B T)]^2} \, dE \ . \tag{2}$$

This equation has been generalized to include a magnetic field by Schotte and Schotte[8] and applied to UBe_{13} by Overhauser and Appel.[9] We assume a Lorentzian form,

$$N(E) = N_0/[1 + (E/\Delta)^2] \ . \tag{3}$$

We first tried to fit the data with $C/T = C_{\text{band}}/T + \beta T^2$ with $N_0 = 22.41$ states/(eV metal-atom spin), corresponding to the entire measured γ. The best fit was obtained with $\beta = 1.04$ mJ (mol)$^{-1}$K^{-4} and $\Delta = 40$ K. This fit was, however, poor compared to that of Eq. (1) and using $\beta = 0.85$, corresponding to a Debye temperature Θ of 213 K,[10] gave a much poorer fit. Very recently, a temperature dependence of Θ below 10 K has been observed.[11] This has little effect on our results. On the other hand, we find that combining Eqs. (1) and (2) allows the data to be fit about as well as with Eq. (1) alone when Δ is larger than about 60 K. The fit parameters were Δ, β^*, and δ, and the optimum values of β^* and δ approached those of Eq. (1) alone for $\Delta > 1000$ K. A magnetic field of 5 T caused practically no change in $C_{\text{band}}(T)$ for $\Delta \geq 60$, consistent with experiment.[12]

We conclude that the $T^3 \ln T$ term in C is really necessary, but also that a peak in $N(E)$ with $\Delta \geq 60$ K cannot be ruled out.[13] Analysis of the fit unfortunately does not provide a unique value for N_0 or, equivalently, γ_0, the band part of γ.

In order to apply the SF theory of UPt_3 in the two-parameter form we have previously used for Pd and $TiBe_2$,[14] we require as input the Stoner factor S and the *many-body* mass enhancement $m^*/m = \gamma/\gamma_0$, $\gamma = \gamma_0(1 + \lambda_{\text{SF}} + \lambda_{\text{ph}})$. Unfortunately, at present neither S nor m^*/m is known for UPt_3. We define S as $\lim q \to 0 \ \omega/q \to 0 \ \chi(q,\omega)/\chi_0(q,\omega)$, where $\chi_0 = 2\mu_B^2 N(E_F)$ is the *band* susceptibility. In the SF theory $S = 1/[1 - N(E_F) I]$, where I is the $q = 0$ limit of the

exchange interaction parameter. In the language of Fermi liquid theory $S=(m^*/m)/(1+F_0^a)$. Other authors[12,15] have defined a related and more directly measurable quantity,

$$\bar{S}=(\pi^2 k_B^2/3\mu_B^2)(\chi/\gamma)=1/(1+F_0^a) \ .$$

The experimental values[12,15] $\bar{S}\simeq 1.5$ for UPt_3 and 1.0 for UBe_{13} yield $F_0^a\simeq -0.3$ and $F^a\simeq 0$, respectively. Since the $T^3\ln T$ term is proportional to $(F_0^a)^3$,[4] these values are consistent with the absence of this term in UBe_{13} and its presence in UPt_3. It is not clear, however, how good a measure of exchange enhancement F_0^a is in a metal. There is a positive electron-phonon contribution to F_0^a which cancels in the susceptibility to leading order in the parameter Debye energy divided by bandwidth. The experimental value of F_0^a is thus an underestimate of the exchange interaction.

In a nearly localized system $S=\bar{S}(m^*/m)$ is large primarily due to the m^*/m factor. In UPt_3, however, F_0^a is rather far from the localization limit of $\simeq -\frac{3}{4}$ (Ref. 16) and the prediction of p-wave pairing of Valls and Tesanovic[17] should be viewed with caution. Of course, F_0^a in UPt_3 is also far from the ferromagnetic limit of -1 and the same applies to the SF results. We believe, however, that the two-parameter SF theory[14] considered as a phenomenological theory has validity for systems that are exchange enhanced but not necessarily in the extreme nearly ferromagnetic limit. The two-parameter theory, where the second parameter describes the q dependence of $I(q)$, is, in fact, not very different from the polarization potential theory,[18] which seems to enjoy a better reputation. In UPt_3 the one-parameter (contact) interaction model may actually not be bad for the tightly bound f electrons.

Recent band-structure calculations[19] yield values for γ_0 [or $N(E_F)$] that seem unreasonably low: $\lambda_{SF}+\lambda_{ph}$ on the order of 20 is required to fit the observed γ. The inclusion of correlation effects U_{ff} could have a large effect on the band-structure result based on the local-density functional approximation (LDF). An attempt has been made recently to attribute the discrepancy between the measured bremsstrahlung isochromat spectroscopy—and the calculated LDF—density of states to such correlations.[20] Since these effects cannot yet be reliably calculated, we are still faced with the problem of estimating λ_0.

In order to obtain consistent values for γ_0 and S, and thus λ_{SF} and λ_{SF}^g, the p-wave pairing parameter, we employ a slightly modified version of the method developed by Boring, Albers, Stewart, and Koelling[21] (BASK) for UAl_2. The input for the calculation is the measured χ, the C/T fit parameters β^*, γ, and δ, and the phonon parameters λ_{ph} and β. These data are fitted to the theoretical results of the paramagnon theory:[22,23]

$$\frac{C}{\gamma_0 T}=1+\lambda_{SF}+\lambda_{ph}+\frac{18\pi^2(S-1)^2}{10S}\left(\frac{T}{T_F}\right)^2\left[\ln\left(\frac{T}{\bar{P}\bar{T}_F}\right)+1.78\right] \ , \quad (4)$$

where $\bar{T}_F=(4/\pi)T_F/(S-1)$ and $E_F=k_B T_F$ is the effective Fermi energy of the f band. Instead of Eq. (A9) of BASK, we have used

$$\lambda_{SF}=(\tfrac{9}{2})\ln\left[1+\frac{\bar{P}^2(S-1)}{12}\right]-(\tfrac{3}{8})\bar{P}^2(S-1)/S \ , \quad (5)$$

where $\bar{P}$ is a cutoff of the paramagnon model.[22] We note that the original paramagnon model is also really a two-parameter theory, although perhaps not as physically satisfying as the q-dependent I model. The second term in Eq. (5) was neglected in Ref. 22 in the limit of large S; its inclusion does not change the results significantly. The equations are solved numerically and yield S, T_F, $\bar{P}$, λ_{SF}, and thus, and this is the nicest feature of the method, m^*/m, γ_0, and $N(E_F)$. Since the input λ_{ph} is not well known, we run the procedure for several values. For UPt_3 we have taken as input $\beta=0.85$ mJ (mol U)$^{-1}$K^{-4}, β^*, γ, and δ as given below Eq. (1), and $\chi=8.5\times10^{-3}$ emu/(mol G). The results are shown in Table I, where λ_{SF}^g was calculated as in Ref. 14 but with a constant exchange parameter $N(E_F)I=(S-1)/S$ and the calculated $\bar{P}$. S-wave pairing is seen to be suppressed unless λ_{ph} is quite large, $\gtrsim 4$. The p-wave interaction is attractive, but we do not want to speculate on the magnitude of the transition temperature, as this quantity is strongly affected by renormalization effects which cannot yet be accurately calculated.[14,24]

We have also applied the method to the SF systems UAl_2 and $TiBe_2$ and the results are shown in Table I. For UAl_2 the inputs are given in BASK and for $TiBe_2$ (Ref. 25) we used $\beta=0.012$ ($T_{Debye}=785$ K), $\beta^*=-0.286$, $\delta=0.094$, $\gamma=51.9$, and $\chi=8.0$ in the same units as above. The significantly larger S for $TiBe_2$ is expected and gives us some confidence in the method. Also, as a consistency check, we have refitted the C/T data assuming a C_{band} term with N_0 adjusted to give the γ_0 from Table I and $\Delta\simeq T_F/2$. New fit parameters β^* and δ were then determined and found to differ little from those used as input.

The numbers in Table I indicate that if UPt_3 is a p-wave superconductor, then $TiBe_2$ and UAl_2 should be also. That they are not may be due to the low residual resistivity ratios of the samples investigated so far. The large mass of UPt_3

TABLE I. Results of the self-consistent determination of the spin-fluctuation and pairing interaction parameters. Units are T_F in K, γ_0 in mJ (mol A)$^{-1}$K^{-2}, $N(E_F)$ in states/(eV A spin). $A=UPt_3$, UAl_2, or $TiBe_2$.

	λ_{ph}	S	T_F	$\bar{P}$	λ_{SF}	γ_0	$N(E_F)$	m^*/m	λ_{SF}^g
UPt_3	0.3	8.5	458	2.10	4.0	73.2	15.5	5.34	0.39
UPt_3	1.0	10.5	596	2.05	4.7	59.3	12.6	6.69	0.52
UAl_2	0.3	10.3	412	1.23	2.9	33.2	7.0	4.24	0.67
UAl_2	1.0	13.6	470	1.20	3.6	25.6	5.4	5.58	0.84
$TiBe_2$	0.2	35.1	8661	0.44	1.9	16.6	3.5	3.10	0.61

makes the ratio of coherence length to mean free path more favorable.[26]

The reasonable results of the BASK procedure show that the phenomenological SF picture at least does not lead to gross inconsistencies. Whether this is merely fortuitous remains to be seen. It is not clear how well such complex systems can be simulated with a narrow-band paramagnon model assuming a parabolic dispersion. In addition, we have not corrected for orbital and diamagnetic contributions to the measured χ, the effect of scattering on the $T^3 \ln T$ term[7] has been neglected, and periodic potential spin-orbit effects are assumed to be effectively lumped into the parameter I. Nevertheless, in view of the lack of real theoretical understanding of the band-structure and many-body effects in UPt_3, we believe it is worthwhile to apply the phenomenological SF theory.[27]

Recently, Oguchi, Freeman, and Crabtree[28] have proposed a two-band model for UPt_3 in which the heavy "f" electrons have a large S and the light "d" electrons have $S \simeq 1$ and are superconducting with conventional s-wave pairing. Our results would apply to the f band. If SF effects in the f band are very strong, their effect on the effective mass and pairing interaction of the d electrons must be investigated. We assume the d electrons to be coupled to the f electron-spin fluctuations through an effective coupling constant I_{df}. It turns out that only the bubble diagrams of the f electron SF contribute. We find directly $\lambda_{SF,d} = (\frac{1}{3})(I_{df}/I_f)^2 \lambda_{SF,f}$, which can be much smaller than $\lambda_{SF,f}$. Thus there may be very little suppression of s-wave superconductivity in the d band by f-band SF. The contribution of the SF bubble diagrams to the pairing interaction is *repulsive* for *both* s- and p-wave pairing, and therefore we see no argument from this mechanism against conventional pairing in the "light" d band. Oguchi *et al.*[28] also suggest that λ_{ph} in the f band could be strongly enhanced by the mechanism proposed by Kim.[29] There is, however, reason to doubt that this is really a large effect.[30]

In summary, we conclude that it is consistent to consider UPt_3 as a spin-fluctuation system. Whether the superconductivity is p or s wave seems to depend on whether the single- or two-band model is more appropriate. This question cannot be answered in the context of the simple models discussed here.

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