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Possibility of Coexistence of Bulk Superconductivity and Spin Fluctuations in UPt$_3$

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Convincing evidence has been discovered for bulk superconductivity in UPt$_3$ at 0.54 K based on specific-heat, resistance, and ac susceptibility measurements. In addition, new evidence is presented that indicates that UPt$_3$ is a spin-fluctuation system. If true, this is the first coexistent superconductor–spin-fluctuation system.

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We have found strong evidence of bulk superconductivity in UPt$_3$ in the course of an investigation to determine if it is a spin-fluctuation system.[1,2] For our specific-heat, resistance, and ac susceptibility measurements, we prepared three batches of flux-grown single crystals and also arc-melted polycrystalline material. In additon, experiments on the effect of annealing these small-mass (~1 mg) needlelike crystals have been performed.

Our first evidence for superconductivity in UPt$_3$ was the resistance curve in Fig. 1(a), showing a drop to zero resistance at about 0.54 K with a transition width of 0.030 K. Then ac susceptibility measurements also indicated that the sample was superconducting, with a diamagnetic transition starting at 0.50 K and a transition width greater than 0.050 K. Annealing improved the transition as shown in Table I which summarizes the results for various samples. We then measured the low-temperature specific heat of both unannealed and annealed crystals from this first batch. The specific heat of the annealed crystals is shown in Fig. 1(b); the data for the unannealed crystals were similar but with a broader transition. The measured specific-heat discontinuity, ΔC, divided by the linear term in the specific heat, γT, is 0.48 at $T = 0.40$ K. As is usual[3] for broad transitions, one may extrapolate C from below 0.40 K upwards to an idealized sharp transition, giving $\Delta C/\gamma T_c > 1.0$. However, it is clear that even the broadened transition in Fig. 1(b) is so large that it must be due to the majority hexagonal, structure type DO19, UPt$_3$ phase present. Metallography cannot detect any second phase in these needlelike single crystals, which is a form that generally exhibits the highest possible phase purity. A conservative upper limit for second phase would be 1%–2%.

The coincidence of transition temperatures measured by three different techniques [$T_c^{\text{onset}}(\rho) = 0.54$ K, $T_c^{\text{onset}}(\chi) = 0.53$ K, and $T_c^{\text{onset}}(C) = 0.54$ K] and the heat-capacity anomaly are clear evi-

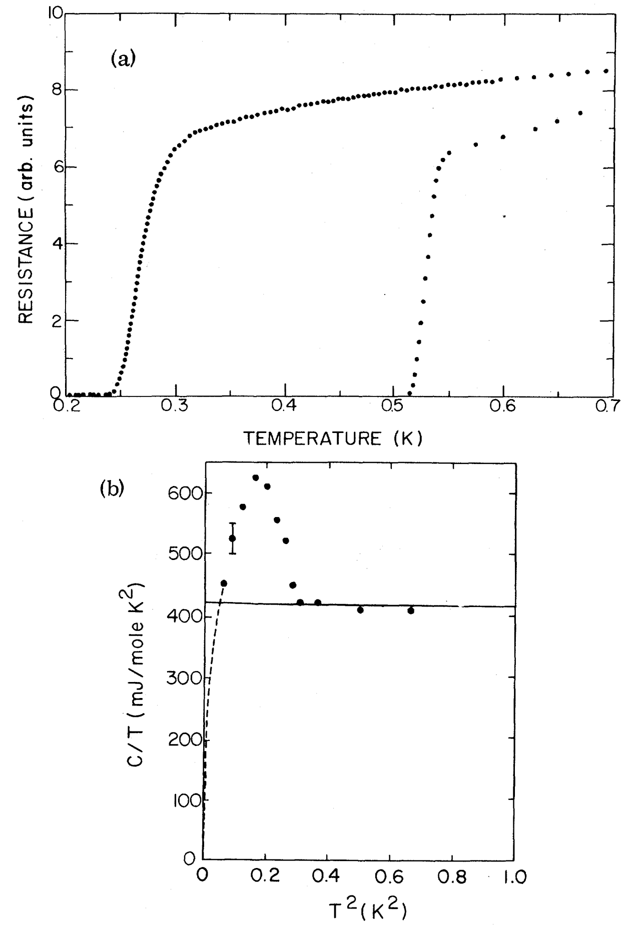

FIG. 1. (a) Resistance vs temperature for samples 1 and 3 of the flux-grown single crystals. Note the lack of any anomaly at 0.54 K in the lower-T_c material. (b) Specific-heat data at lower temperatures on annealed (1200 °C, 12 h) UPt$_3$ crystals, batch 1. The dashed line shown is a reasonable extrapolation of the data to $T = 0$ which achieves entropy balance at T_c. The low-temperature data are shifted about 5% from the high-temperature extrapolation shown in Fig. 2. Since data on unannealed crystals (not shown) agreed with the data shown on annealed crystals above T_c, this difference is thought to be a systematic error between the two platforms used, with the higher-temperature platform having the better absolute accuracy (± 3%). The precision of the data from both platforms is better than 2%.

TABLE I. Transition temperature, T_c, and width, ΔT_c, for the various preparations of UPt$_3$. The first three samples were grown from Bi flux.

	Resistive $T_c^{\text{onset}}/\Delta T_c$ (K/K)	χ_{ac} $T_c^{\text{onset}}/\Delta T_c$ (K/K)
Sample 1	0.54/0.030	0.50/> 0.050
Sample 1 annealed		0.53/0.030 [a]
Sample 2	0.52/0.020	
Sample 3	0.29/0.040	
Sample 3 annealed		0.51/0.020
Sample 4 (arc-melted)		0.40
Sample 4 annealed		0.49/0.080
Czochralski grown crystal[b]	0.5	

[a] This sample had a broad tail to lower temperatures.
[b] J. J. M. Franse, private communication.

dence of bulk superconductivity in UPt$_3$. In order to make this abundantly clear, we discuss (and discard) here the possibility of the ΔC arising from another type of transition, such as itinerant antiferromagnetism, while the resistive and inductive anomalies were due to a tiny amount of a superconducting second phase with coincidentally the identical T_c. This can be ruled out for two reasons. First, as seen in Table I, the T_c's for the arc-melted and Czochralski preparations (which contain no bismuth flux) agree with the samples grown out of the bismuth solvent. The differing conditions for the three types of preparation would suggest that the same second phase could not occur in all of them. Second, there is a range of T_c's shown in Table I for various batches of single crystals. In the cases where $T_c < 0.54$ K, there is *no resistive anomaly* to better than 1% [see Fig. 1(a)] present above the measured T_c, i.e., there is no anomaly at 0.54 K. If indeed the majority phase of UPt$_3$ had some transition at 0.54 K which gave the specific-heat anomaly in Fig. 1(b), it is extremely unlikely that, in the other batches, the (supposedly) nonsuperconducting transition in the majority phase would also (again coincidentally) occur at the lower, resistive T_c or that it would give no resistive anomaly if it remained at 0.54 K. Therefore, UPt$_3$ is a bulk superconductor with a superconducting T_c as high as 0.54 K.

We have discovered new evidence that indicates that if, as is generally accepted,[4-8] UAl$_2$ or TiBe$_2$ are spin-fluctuation systems, then UPt$_3$ is also. This new evidence consists of two parts. First, the specific heat in zero magnetic field is plotted in Fig. 2. The line shown through the data is a least-squares computer fit by

$$C = \gamma T + \beta T^3 + C_{\text{SF}}, \qquad (1)$$

where

$$C_{\text{SF}} = \gamma^* T + \delta T^3 \ln T/T_{\text{SF}}.$$

This spin-fluctuation contribution to C was first calculated by Doniach and Engelsberg[9] for ^{3}He. Although one cannot quantitatively analyze the results of a fit to Eq. (1), derived for ^{3}He, of data for a material with a nonspherical Fermi surface and highly correlated electrons, the fact that the data fit a $\gamma' T + \beta T^3 + T^3 \ln T/T_{\text{SF}}$ dependence [and not $\gamma' T + \beta T^3$ plus either a $1/T^2$ (Schottky) or a $1/T$ (spin glass) dependence—see Fig. 2] is certain-

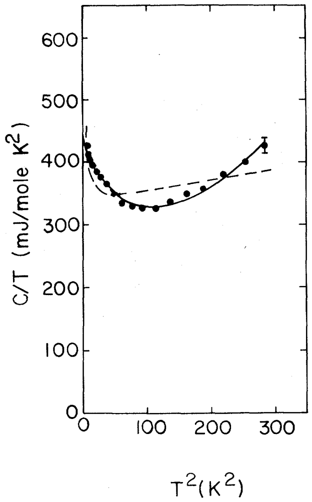

FIG. 2. Higher-temperature specific-heat data for unannealed single crystals, batch 1, of UPt$_3$. The line through the data is a fit by Eq. (1); the dashed line is the same fit but with $1/T^2$ replacing the $T^3 \ln T/T_{\text{SF}}$ term. A $\gamma T + \beta T^3 + 1/T$ fit lies within 2% of the dashed curve at all temperatures. Clearly, the added $T^3 \ln T/T_{\text{SF}}$ term gives the best fit to the specific-heat data, as is true for UAl$_2$ and TiBe$_2$.

ly an indication[10] that the material is a spin-fluctuation system. The fit by Eq. (1) for the UPt$_3$ specific-heat data shown in Fig. 2 is as good as similar fits[4, 5] for UAl$_2$ and TiBe$_2$. Previous measurements of an enhanced magnetic susceptibility at 4 K ($\chi = 7.0\times10^{-3}$ emu/mole in Ref. 1 and similarly in Ref. 2) are another, quite strong, indication that UPt$_3$ is a paramagnon, or spin-fluctuation, system. Hence, the cases for spin fluctuations in UPt$_3$, TiBe$_2$, and UAl$_2$ are identical. Another new piece of evidence that UPt$_3$ is a spin fluctuator is that the temperature depedence of the resistance near 0.5 K is quite close to T^2 as predicted for spin fluctuators by Doniach[11] and as found[12] for UAl$_2$.

We would therefore conclude that UPt$_3$ is a bulk superconductor with strong indications that it is also a spin-fluctuation system. We would like to stress that, if this conclusion is borne out by other experiments, then UPt$_3$ is unique, as there are *no* other known coexistent superconductor-spin-fluctuation systems. Further, although UPt$_3$ has an enormous γ (see Fig. 2) of 450 mJ/(g-atom U)$\cdot$K^2, it is clearly different from the other two known "heavy fermion" superconductors, CeCu$_2$Si$_2$[13] [$\gamma = 650$–1300 mJ/(g-atom Ce)$\cdot$K^2] and UBe$_{13}$[14] [$\gamma \sim 1100$ mJ/(g-atom U)$\cdot$K^2]. This difference is not just the $T^3 \ln T$ term in C (which does *not*[15] work for the upturns in UBe$_{13}$ and CeCu$_2$Si$_2$) and the large enhanced susceptiblity[1, 2] (χ/γ is 4 times larger for UPt$_3$ than for the other two), but is most dramatically shown by the curve of resistance, R, versus temperature in Fig. 3. While both of the "heavy fermion" superconductors have curves that are quite similar, UPt$_3$ is completely different.

Finally, as a *pure* speculation note that paramagnons, although harmful to normal BCS superconductivity, are predicted[16] to enhance triplet, or p-wave superconductivity. Theory also predicts[17] that impurity scattering has a severe effect on p-wave superconductivity. UPt$_3$ shows unusual defect sensitivity of T_c in two respects. First, the fact that T_c falls from 0.54 to 0.27 K with a change in $R(300\text{ K})/R(T_c^{+})$ of from 145 to 43 is a severe dependence of T_c on resistance ratio. Second (and this is again unique), we know of no other homogeneous, bulk superconductor whose T_c is totally suppressed ($T_c < 0.050$ K) by grinding. (T_c recovers upon annealing, i.e., this is truly a bulk and not a second-phase effect.)

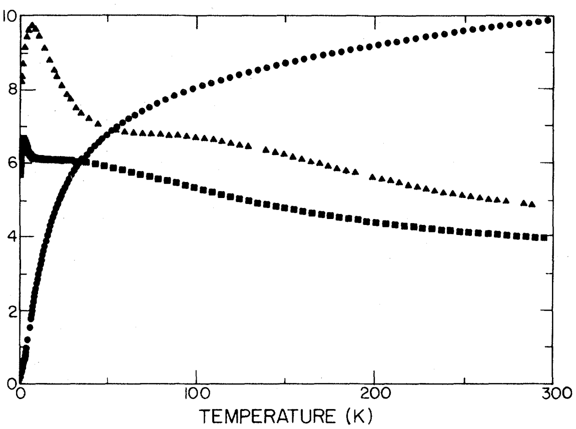

FIG. 3. Resistance vs temperature of CeCu$_2$Si$_2$ (triangles), UBe$_{13}$ (squares), and UPt$_3$ (dots). Note the similarity of the data for the first two—both have low-temperature peaks and shoulders at higher temperature.

While the above discussion is consistent with UPt$_3$ being a p-wave superconductor, this is clearly mere speculation and must be investigated by further measurements.

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