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Sample dependence of the spin-glass behavior in UPt_3

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We present measurements of the recently reported spin-glass behavior in UPt_3 via zero-field-cooled vs field-cooled magnetic susceptibility and remanent magnetization data on a wide variety of pure UPt_3 samples. These samples include a float zone-method single crystal (unannealed and annealed), needle single crystals from arc-melted samples, neutron-irradiated (10^{18} n/cm^2 and 10^{19} n/cm^2) polycrystalline material, and also $UPt_{3\pm x}$ material. The jump in the specific heat at the superconducting transition, $\Delta C(T_c)$, in several samples is discussed, with the result that, contrary to previous work on polycrystalline material, the largest $\Delta C(T_c)$ value in single crystal UPt_3 is found in the sample with the *largest* spin-glass effect. [S0163-1829(97)02125-5]

I. INTRODUCTION

Recently, via doping experiments on UPt_3, it was discovered[1] that both doped and pure UPt_3 display the classic signs of a spin glass: deviations between zero-field-cooled (ZFC) and field-cooled (FC) dc magnetic susceptibility below a temperature T_{freezing}, a time-dependent remanent magnetization, and a peak in the ac magnetic susceptibility with a small ($<0.05\ T_{\mathrm{peak}}$) change over a decade of frequency. We report here on an extended study of the spin-glass behavior in pure UPt_3 in a large variety of samples, including both float zone and needle single crystals, neutron-irradiated polycrystalline samples, and polycrystalline samples as a function of stoichiometry, i.e., $UPt_{3\pm x}$. The primary measurement techniques employed to characterize the spin-glass properties were ZFC-FC χ_{dc} and the remanent magnetization directly after the field was set to zero, while specific-heat measurements were used to characterize the superconductivity.

II. EXPERIMENT

The float-zone single crystal measured was a piece from a large sample produced by the float-zone method. One piece of the crystal was annealed in high (10^{-9} mbar) vacuum (1280 °C for 3 h, followed by cooling over 2 h to 900 °C, followed by cooling to room temperature over 12 h) so that a comparison between the spin-glass properties before and after annealing could be carried out.

The whisker, or needle single crystals, were ''harvested'' from repeated arc-melting of high-purity stoichiometric $UPt_{3.00}$, using 99.998% pure Pt from Johnson-Matthey Aesar and electrotransport refined U from Ames Laboratory. Such crystals spring out from the upper surface of the arc-melted beads as they cool through the melting temperature. In addition, we attempted with some success to produce needle crystals from beads of $UPt_{3.04}$ (nominal) in order to attempt to alter the stoichiometry (and therefore the ZFC-FC properties) of the crystals. This led to the discovery that excess Pt in the bead severely hinders the production of the crystals. An electron microprobe study of both the float-zone single crystal and of the starting beads of both the $UPt_{3.00}$ and $UPt_{3.04}$ samples was carried out using a JEOL superprobe model 733, taking 30 separate regions and measuring each for 100 sec, to determine the stoichiometry.

The polycrystalline UPt_3 samples for neutron irradiation have already been previously characterized via inductive measurements of T_c and the specific heat,[2] superconducting

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TABLE I. Parameters for various pure UPt_3 samples.

	$\frac{\chi_{FC}-\chi_{ZFC}}{\chi_{ZFC}}$	$T_f(K)$	Remanent magnetization[a]	$\frac{\Delta C}{T_c}$(mJ/mole K^2)	$T_c^{mid}(K)$
Unannealed float-zone crystal					
$H\|a$-b	0.041	16±2	0.044	200	0.43
$H\|c$	0.26	16±2	0.109		
Annealed float-zone crystal					
$H\|a$-b	0.11	16±2	0.046	180 (250[f])	0.465
$H\|c$	0.45	16±2	0.216		
Newly prepared needle crystals					
$H\|a$-b	0.15	55±5		9.0	0.48
$H\|c$	0.30	55±5	0.036		
Four-year-old needle crystals					
$H\|a$-b	0.115	55±5	[e]	10	0.47
$H\|c$	0.14[b]	55±5			
Unirradiated polycrystalline	0.043–0.057[c]	55±5	0.0048–0.0057	24	~0.4
10^{18} n/cm^2	0.043–0.092[c]	55±5			
10^{19} n/cm^2	0.012–0.016[c]	45±10	0.0041		
$UPt_{3.00}$	0.008	~8	[e]	45	0.28
$UPt_{3.04}$	0.002	?	[e]	150	0.46
$UPt_{3.04}$[g]	0.007[d]	?	[e]	60	0.50
$UPt_{2.96}$	0.15–0.24	16±2	0.040		
$UPt_{2.96}$ ground	0.23	18±2	0.017		

[a] Remanent magnetization is expressed as $\chi(H=0)/\chi(200\ \mathrm{G})$ at 0.6 T_f, where $\chi(H=0)$ is measured about 3 1/2 minutes after $\chi(200\ \mathrm{G})$ is measured, following the procedure in the text.

[b] Since the newly prepared crystals were prepared from one of the original beads used to produce these four-year-old crystals, this smaller value may indicate an aging effect, but more work needs to done before any definitive statement.

[c] Polycrystalline samples, which display some preferential orientation, were measured in two orthogonal directions.

[d] Values are generally reduced by half via annealing.

[e] Value too small to measure reliably.

[f] Measured to the second, lower peak.

[g] This is the sample which was ground and shown in Fig. 6.

transition temperatures were 0.50, 0.20, and 0.08 K for the unirradiated, 10^{18} n/cm^2, and 10^{19} n/cm^2 ($E>1$ MeV) samples, respectively, where n/cm^2 is the unit of fluence, neutrons/cm^2 total irradiation.

Polycrystalline $UPt_{2.96}$, $UPt_{3.00}$, and $UPt_{3.04}$ were prepared using normal arc-melting techniques, using good quality ~99.95% U from Cameco and 99.998 Pt from Johnson Matthey Aesar.

The specific heat was measured using the time-constant method.[3] Magnetic measurements were made in a Quantum Design Squid magnetometer. (Demagnetizing corrections were estimated for all measurements to be less than or equal to 6%, and were not taken into account.) For the ZFC-FC data, the field used was 200 G. It was found that the zero-field-cooled data for the float-zone single-crystal samples with the field in the c-axis direction are *extremely* sensitive to the exact field in which the samples are cooled. A remanent field of only ±1 G can double (−) or halve (+) the difference between the ZFC data and the data measured in +200 G. For the remanent magnetization, the samples were cooled in a 200 G field to 10 K from room temperature, where (at 10 K) the samples were held for 15 min. Since 10 K is below T_{freezing}, but still a significant fraction thereof, this procedure helps to maximize the alignment of the random spins. Samples were then measured, after which the field was ramped down to zero and χ_{dc} was again measured. The same computer program, with the same time between steps, was used for all the samples measured so that, although the absolute values—due to the strong time dependence involved—are rather arbitrary, the intercomparison between samples offers a correct relative estimate.

III. RESULTS AND DISCUSSION

A. Single crystals

The χ_{ZFC}-χ_{FC} results, expressed as a fraction of χ_{ZFC} are given in Table I. The first result to remark on is that the random spins tend, in the small 200 G field, to show increased alignment (vs zero-field cooled) when cooled in field primarily in the c-axis direction, whereby the difference with the field in the a-b plane between χ_{dc}(ZFC) and χ_{dc}(FC) is markedly smaller. (This is equivalent to saying that the response of the U 5f spins in the $H\|c$-axis direction tends more to being frozen at 1.8 K—the spins do not respond to the 200 G field applied after reaching 1.8 K.)

This difference in $(\chi_{\mathrm{FC}}$-$\chi_{\mathrm{ZFC}})/\chi_{\mathrm{ZFC}}$ for the two field directions is clearly the case (Table I) for the annealed float-zone crystal (shown in Fig. 1), for the unannealed float-zone crystal, and for the recently made whisker crystals (see Fig. 2)

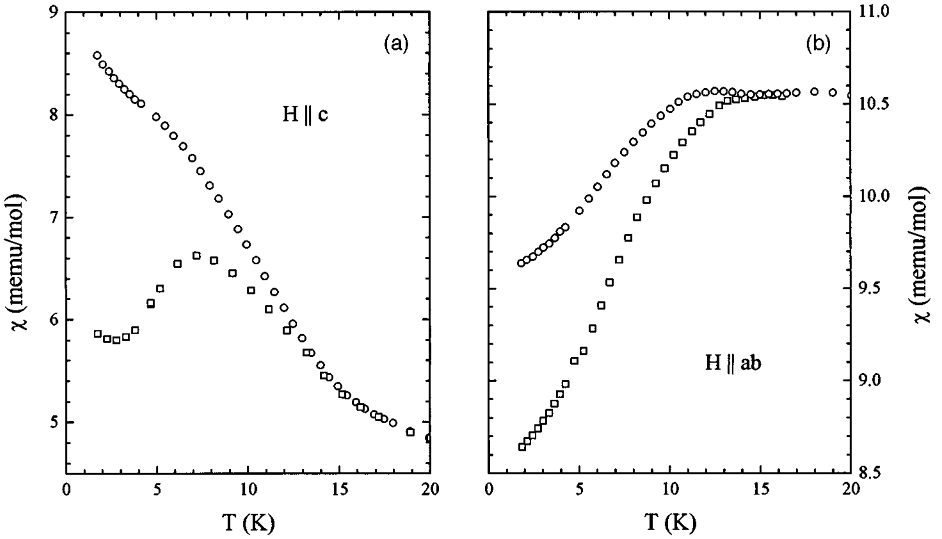

FIG. 1. (a) and (b) Magnetic susceptibility, χ, vs temperature for a float-zone method single crystal cooled in zero (± 0.2 G) field to 1.8 K and then measured in 200 G as a function of increasing temperature (squares), as well as measured in field (200 G) (circles) while cooling from 30 K. The two field directions with respect to the crystal axes were determined approximately using the known direction dependence of χ. The third orthogonal direction gave results within 2% of the $H \| a,b$ results, indicating good alignment. Note the extremely large difference between the field-cooled (FC) and zero-field-cooled (ZFC) data for $H \| c$, as well as the distinct peak in χ_{ZFC} for $H \| c$.

while the difference between the c-axis and the a-b plane for the 4 year-old whisker crystals is, while still observable, quantitatively smaller. The effect is far and away the largest for the annealed float-zone single crystal (see Fig. 1 and Table I). What is further remarkable to note is that in this sample, field-cooled data in the c direction reach the value observed for the ZFC a-b plane susceptibility.

Although these $(\chi_{FC}$-$\chi_{ZFC})/\chi_{ZFC}$ results seem quite definitive for differentiating the size of the spin-glass effect among the various UPt_3 samples, the size of the differences involves not only the number of spins involved, but also their freedom to reorient. In order to provide a comparison method for characterizing the spin-glass behavior in UPt_3, the remanent magnetization of the samples was also measured, and is shown in Table I. These values, which are also only indirect measures of the microscopic behavior of the spins, at least seem to roughly scale with the $(\chi_{FC}$-$\chi_{ZFC})/\chi_{ZFC}$ numbers. Thus (Table I), the remanent magnetization in the annealed float-zone crystal, $H \| c$, is a factor of 4.7 larger than for $H \| a$-b, while the ratio of the respective $(\chi_{FC}$-$\chi_{ZFC})/\chi_{ZFC}$ values is 4.1. Similar comparisons hold for most of the other samples, although, since it is technically easier to measure small differences in χ_{FC} and χ_{ZFC} than it is to measure small remanent magnetizations that lie near the resolution limit of the Quantum Design susceptometer, some scatter in the remanent magnetization values in samples where the spin-glass effect is small is unavoidable. (It is worth noting that the remanent magnetization values, as characteristic of spin glasses, decay with time as a function of log [time].)

By examining the χ data for the annealed float-zone crystal and the needle crystals (Figs. 1 and 2), the following comparisons can be made: (1) The float-zone crystal shows, in addition to the already known[4] peak at 19 K in χ in the field parallel to the a-b plane direction, a peak in χ_{ZFC} for $H \| c$ at 7 K. This was not seen in earlier, presumably field-cooled, data,[4] and is also not, within the scatter, apparent in the needle crystal data shown in Fig. 2. (2) The freezing temperatures, T_f (listed in Table I), which may be estimated[5] by where the ZFC and FC curves join (see, e.g., Figs. 1 and 2), are radically different in the two types of crystals ($T_f \simeq 16$ K for annealed float zone, which is, within a $\simeq 2$ K error bar, the same as for the unannealed float-zone crystal, while $T_f \simeq 55$ K for the newly produced crystals (see Fig. 2), similar to the value for the 4-year-old whisker crystals). (In order that the remanent magnetization measurements on samples with such differing T_f values be comparable, the remanent magnetization for these high T_f samples were measured at 35 K, the same fraction of T_f as used for the low T_f samples.) This radical difference in T_f indicates a far stronger resistance to spin reorientation in the needle crystals than in the float-zone crystals, independent of annealing. This stronger ''glassy'' character is presumably dependent on the type and distribution of the defects that cause local U spins not to be fully compensated. In the polycrystalline specimens reported in Ref. 1, including the ground specimen, T_f was in the range 10–20 K. (Note, however, the data on the 9-year-old polycrystalline sample[6] in Table I.) Thus, the rapid process, with the accompanying rapid cooling, by which the needle crystals are extruded from the surface of the cooling arc-melted bead appears to be more important for the spin-glass properties than the difference between single crystal and polycrystalline material. (3) In the FC curves in Fig. 2 there is an upturn in χ below 5 K in both field directions for the needle crystals that is not present in the float-zone crystals nor in the typical polycrystalline material. (The UPt_3 sample for irradiation, see below, with its similar T_f, does however show such an upturn.) Although the needle crystals are made with much higher purity material than the float-zone crystals, such an upturn seems reminiscent of an impurity.

B. Polycrystalline samples: Comparison

Turning now to a discussion of the neutron irradiated polycrystalline UPt_3 sample,[2] Figs. 3–5 and Table I show the

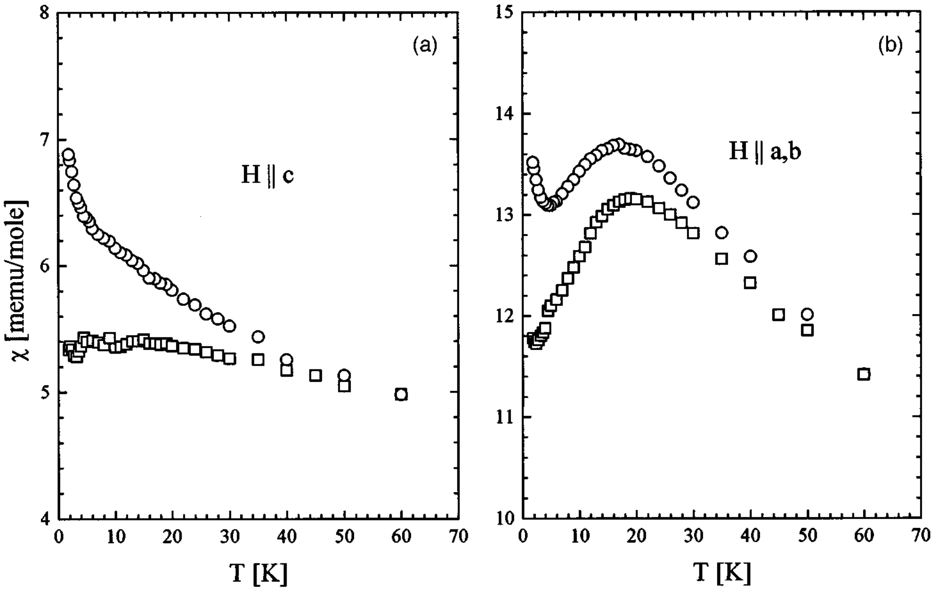

FIG. 2. (a) and (b) Magnetic susceptibility, FC (200 G) (circles) and ZFC (squares), vs temperature for $H \| c$ and H in the a,b plane for recently made UPt_3 whisker crystals. Since the needle crystals have the c axis aligned along the needle axis, the alignment is easy to achieve. Note that T_{freezing}, which is approximately where the ZFC and FC curves diverge, is much larger ($\sim$55 vs $\sim$16 K) for the needle crystals than for the float-zone crystal, Fig. 1.

interesting result that damage induced by 10^{19} n/cm^2 neutron irradiation actually suppresses the ZFC-FC difference in χ_{dc}. This is contrary to what one might expect, i.e., more damage implies more defects and defects are responsible for the uncompensated U spins and therefore the spin-glass behavior. As reported in Ref. 2, while 10^{18} n/cm^2 changes, as measured by the specific heat, the spin fluctuation temperature only slightly, 10^{19} n/cm^2 essentially destroys the spin fluctuations in UPt_3. Thus, the magnetic behavior evidenced by the spin fluctuations may be linked to the spin-glass behavior in UPt_3.

Another kind of defect that is possible to readily bring about is the effect of stoichiometry. Although polycrystalline $UPt_{3.04}$ and $UPt_{3.00}$ seem to have $(\chi_{\mathrm{FC}}\text{-}\chi_{\mathrm{ZFC}})/\chi_{\mathrm{ZFC}}$ values of 1.5% or less [grinding changes this to 6% (Ref. 1)], $UPt_{2.96}$ (see Table I) shows a significantly larger difference ($\sim$20%), larger than the effect from grinding. In fact, this result for the substoichiometric $UPt_{2.96}$ is quantitatively larger than for any polycrystalline stoichiometric sample measured and seems comparable to the single-crystal results, with the exception of $H \| c$ for the float-zone method crystal.

This raises the question, is the large $(\chi_{\mathrm{FC}}\text{-}\chi_{\mathrm{ZFC}})/\chi_{\mathrm{ZFC}}$ result (or, as stated above, this tendency of the spins to be frozen at low temperatures at low field) in $UPt_{2.96}$ an indication that the single crystals are also substoichiometric in Pt? We have performed electron microprobe measurements on the unannealed float-zone crystal and on a slice of an arc-melted button of both $UPt_{3.00}$ and $UPt_{3.04}$. All three samples give a stoichiometry of $UPt_{2.958\pm0.01}$, with the only deviation being that for the $UPt_{3.04}$ sample the 1 μm wide electron beam found 3 of the 30 regions to be Pt rich, with an average stoichiometry of $UPt_{3.4}$. This is an indication of the presence of a small amount of second phase (UPt_5) being present in the Pt-rich ($UPt_{3.04}$) sample, and that the phase width of the UPt_3 compound is quite narrow in stoichiometry.[7] (The fact that the absolute value of the microprobe results, $UPt_{2.958\pm.01}$, is outside of the error bar for the nominal 3:1 composition may be just a calibration error.) These results show that the float-zone single crystal has in fact the same stoichiometry as the $UPt_{3.00}$ arc-melted button (which has, see Table I, a small FC-ZFC difference). Thus, the explanation for the large spin-glass effect in the crystals is *not* due to a deficiency in Pt, i.e., the fact that $UPt_{2.96}$ also has a large spin-glass effect is not the explanation.

In order to further investigate this large spin-glass effect, an investigation of crystals with increased Pt content was undertaken. The method used was to try to produce needle crystals from arc-melted buttons of composition $UPt_{3.04}$ to see if the spin-glass properties of the crystals can be influenced by the stoichiometry of the button. What was immediately observed is that the needles tended to be produced far less often and are far smaller, indicating that stoichiometry of the bead is an important parameter for the production of the needle crystals. Measurements of the crystals gave results similar to those of the newly prepared (from $UPt_{3.00}$) needle crystals shown in Table I; T_f remains at 55±5 K. Due to the

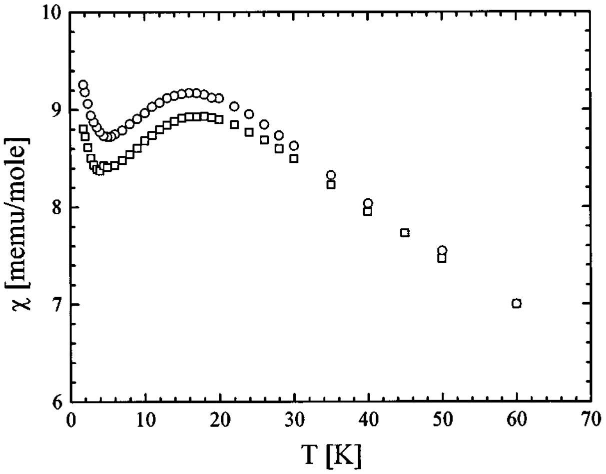

FIG. 3. Magnetic susceptibility vs temperature, FC (200 G) (circles) and ZFC (squares), of the polycrystalline UPt_3 rod used for the neutron irradiation in Ref. 2, with H along the rod axis (see Table I for the slight orientation dependence.) Note that both the ZFC and FC data show a strong upturn below 5 K, compare Fig. 2(b) for the FC needle crystals.

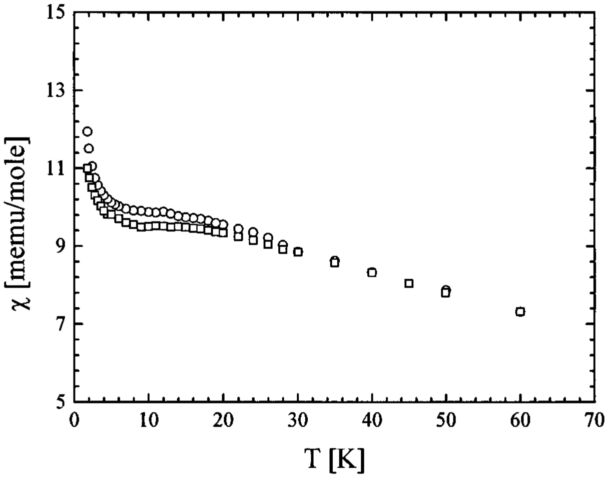

FIG. 4. Magnetic susceptibility vs temperature, ZFC (squares) and FC (circles) (200 G), *H*‖rod axis, for polycrystalline UPt_3 irradiated with 10^{18} n/cm^2. These data are quite similar to those in Fig. 3 for the unirradiated sample in terms of the ZFC-FC deviation, i.e., the spin-glass properties. The magnitude of χ has increased ~20% with the irradiation.

difficulty in polishing a flat surface on the 10μ needle crystals for electron microprobe measurements, the actual stoichiometry of these crystals has not yet been successfully measured.

C. Spin-glass-like behavior and superconductivity

What relationship does this weak magnetic behavior in UPt_3 have to the unusual superconductivity? In Ref. 1, the conclusion was that, for polycrystalline samples, there seemed to be a correlation, best exemplified by the results due to grinding: grinding was reported[8] to destroy bulk superconductivity in UPt_3, and it was found in Ref. 1 that grinding of a polycrystalline specimen strongly increased $(\chi_{FC}$-$\chi_{ZFC})/\chi_{ZFC}$ (from 0.2 to 6 %). However, in the present work we find much larger $(\chi_{FC}$-$\chi_{ZFC})/\chi_{ZFC}$ values in single crystals, as high as 45%. This apparent inconsistency caused us in the present work to measure a ground sample (in an agate mortar to a mesh size of ~250 or ~60μ particle diameter) of polycrystalline $UPt_{3.04}$ for superconductivity via χ_{ac} down to 0.050 K, i.e., to check the result of Ref. 8, where grinding of needle crystals grown from a Bi flux was reported to give no superconducting transition in χ_{ac} down to 0.050 K. The result is shown in Fig. 6. The superconducting transition at 0.48 K is broad, and is only about 10% of the size of the diamagnetic signal seen in a bulk UPt_3 sample. Thus, the correlation put forward in Ref. 1, that the increased value of $(\chi_{FC}$-$\chi_{ZFC})/\chi_{ZFC}$ in ground powder correlates with a disappearance of superconductivity, is indeed substantially correct for this polycrystalline sample.

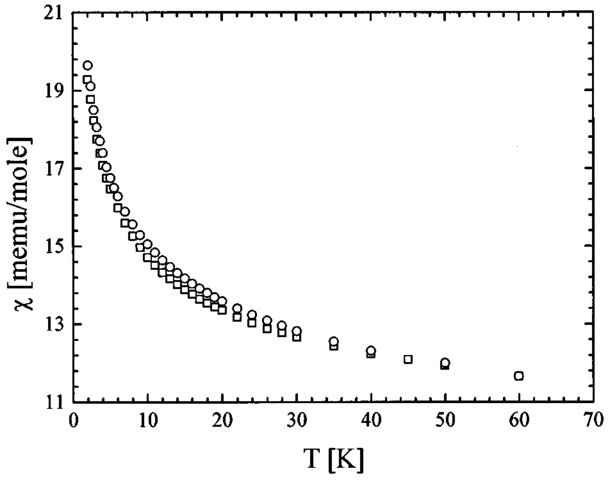

FIG. 5. Magnetic susceptibility vs temperature, ZFC (squares) and FC (circles) (200 G), *H*‖rod axis, for polycrystalline UPt_3 irradiated with 10^{19} n/cm^2. The difference (see Table I) in the FC and ZFC curves has decreased with this heavy irradiation, as well as changing the temperature dependence of χ drastically.

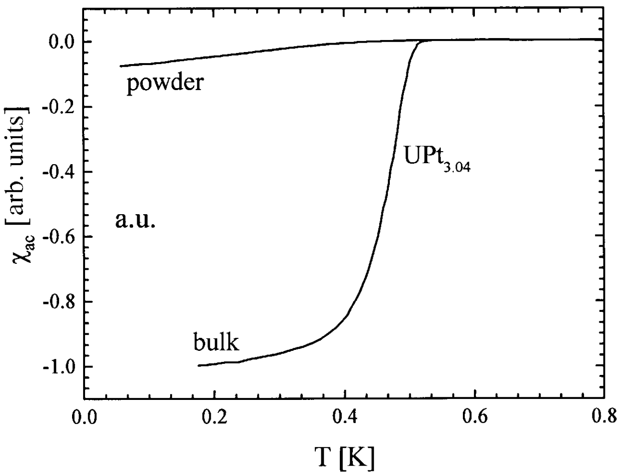

FIG. 6. ac magnetic susceptibility vs temperature of ground $UPt_{3.04}$, showing a broad superconducting transition below 0.5 K, and bulk $UPt_{3.04}$, which shows a full transition.

However, as may be seen from Table I, the sample expected to be the best superconductor, the annealed single crystal, shows the largest spin-glass effect in the present work. In order to investigate the superconducting properties on these specific samples, measurements of the jump in the specific heat, ΔC, at the superconducting transition temperature, T_c, were performed to allow a good determination of how the superconductivity on a bulk scale correlates with changes in the spin-glass properties. These results, for several samples, are shown in the right two columns of Table I; the specific-heat data for the annealed and unannealed float-zone crystals are shown in Fig. 7. One sees from Fig. 7 immediately that the annealed, as well as the unannealed, single crystals are quite good (large ΔC) bulk superconductors. This leads to the inescapable result that the conclusion in our previous work,[1] i.e., that the spin-glass behavior in UPt_3 was the determining, heretofor hidden (deleterious), parameter for superconductivity, is still missing a key variable. In order to try to further determine this variable, or at least to limit the possibilities therefore, let us consider the specific heats of several further samples. The specific-heat data for a good polycrystalline sample [$UPt_{3.04}$, $(\chi_{FC}$-$\chi_{ZFC})/\chi_{ZFC}=0.002$] and a polycrystalline sample [$UPt_{3.00}$, $(\chi_{FC}$-$\chi_{ZFC})/\chi_{ZFC}=0.008$] with a depressed superconducting transition temperature, T_c, and specific-heat jump, ΔC, are

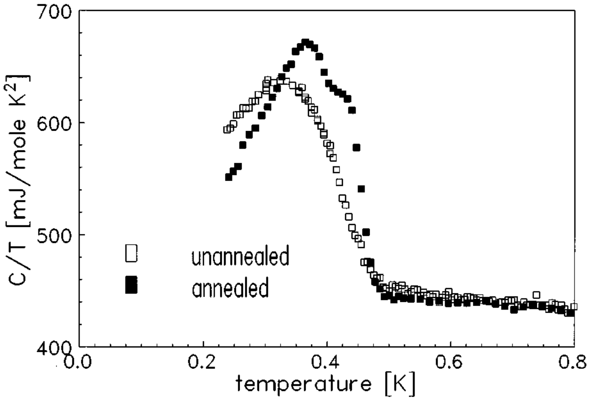

FIG. 7. Specific heat divided by temperature vs temperature of the annealed and unannealed float-zone UPt_3 single crystals. Note the double-peak structure for the annealed sample.

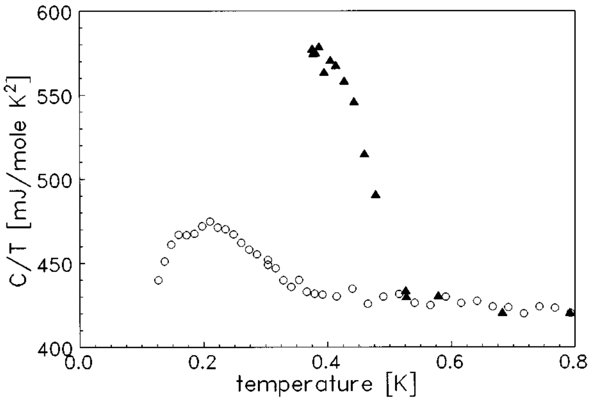

FIG. 8. Specific heat divided by temperature vs temperature of polycrystalline $UPt_{3.04}$ (filled triangles) and $UPt_{3.00}$ (open circles).

shown in Fig. 8. These data, also shown numerically in Table I, serve to further emphasize the dichotomy between the float-zone crystal and all other samples. For the other samples, including the new and old needle crystals and several polycrystalline samples, the correlation between the relative strength of the spin-glass behavior [measured either by $(\chi_{FC}-\chi_{ZFC})/\chi_{ZFC}$ or via the size of the remanent magnetization] and depressed ΔC at T_c, see Table I, continues to be obtained. Only for the float-zone crystal is this correlation exactly the opposite. More work on other float-zone crystals is now underway to try to resolve this conflict.

IV. CONCLUSIONS

The spin-glass behavior, as measured by both the field-cooled vs zero-field-cooled χ_{dc} difference and by the (time-dependent) remanent magnetization, has been measured on a wide variety of polycrystalline and single crystal UPt_3. The samples which were formed by rapid cooling (the needle crystals and the cast rod for the irradiation experiment) show a freezing temperature three times (55 K vs 16 K) that of the other samples.

Concerning the relationship of the spin-glass behavior to the superconductivity, since the discovery[8] of superconductivity in UPt_3, with its coexistent spin-fluctuation behavior, it has often been proposed[8–10] that the superconductivity in UPt_3 is of an unconventional, non-BCS type. If of a non-s-wave nature, the pairing mechanism of the superconducting electrons would be particularly defect sensitive. This is consistent with the wide range of T_c's observed for nominally equivalent samples of UPt_3 and with the wide range of $\Delta C(T_c)$ values observed here (see Table I). If the spin-glass behavior (which has been shown here to be directionally dependent in the single-crystal results) *is* intertwined with the superconducting pairing mechanism, then the large range of sample dependence of the superconductivity observed (see, e.g., Ref. 11) is actually expected due to the known[12] sensitivity of spin-glass behavior to defects, coupled with the defect-susceptible nature of the DO 19 UPt_3 structure.[13] This coupling of spin-glass behavior with unusual superconductivity in UPt_3, if in fact the case, makes, however, for extreme difficulty in making definitive statements, as demonstrated here by the dichotomy between our results for the spin-glass behavior vis a vis superconductivity in a float-zone crystal and our results for polycrystalline and needle crystal samples.

Work is underway to further investigate the extreme sample dependence of superconductivity in UPt_3 in light of the possible linkage to spin-glass behavior.

ACKNOWLEDGMENT

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