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Effect of pressure on spin fluctuations and superconductivity in heavy-fermion UPt_3

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We have determined the effect of hydrostatic pressure on the susceptibility, on the T^2 temperature dependence of the spin-fluctuation resistivity, and on superconductivity in UPt_3. The spin-fluctuation temperature T_s, derived from the slope of resistivity versus T^2, is used within a Fermi-liquid picture to calculate the susceptibility χ at $T=0$ K. The depression of this calculated χ with pressure agrees with the directly measured value $\partial \ln\chi/\partial P = -24$ Mbar^{-1}. Both the superconducting transition temperature T_c and the initial slope of the upper critical field also decrease under pressure. We find that $\partial \ln T_c/\partial P = -25$ Mbar^{-1} and speculate upon correlations between χ and T_c.

In the uranium-platinum series superconductivity occurs[1-3] in the compound that shows the most pronounced spin-fluctuation phenomena, UPt_3. The neighboring phases, UPt_2 and UPt_5, exhibit weaker spin-fluctuation effects in specific-heat[4] and resistivity[5] measurements and do not show, so far, any sign of a superconducting state. UPt_3 can be classified as a heavy-fermion superconductor, i.e., a system that behaves as a Fermi liquid with a large effective mass ($m \simeq 200 m_e$), like $CeCu_2Si_2$ (Ref. 6) and UBe_{13}.[7] The unusual coexistence of spin fluctuations and superconductivity has led Stewart, Fisk, Willis, and Smith[1] to speculate on p-wave superconductivity in UPt_3, in analogy to ^3He. Therefore, it is an intriguing question whether spin fluctuations and superconductivity are intimately connected in UPt_3.

High-pressure experiments on the resistivity of UPt_3 have been performed by De Visser, Franse, and Menovsky[5] and by Wire, Thompson, and Fisk[8] at pressures up to 4.2 and 18 kbar, respectively. De Visser *et al.* pointed out that only at approaching 1.4 K from higher temperatures is a T^2 dependence observed, as predicted by spin-fluctuation theories.[9] The coefficient of this term and its pressure dependence are determined in their analysis by the slope of the ρ vs T^2 curve at the lowest temperatures. For current directions in either the basal plane or along the hexagonal axis, a substantial depression of this coefficient at 4.2 kbar is observed. From both experiments one must conclude that a strong depression of the spin-fluctuation contribution to the resistivity takes place at low temperature.

To examine in greater detail the interplay of spin fluctuations and superconductivity in UPt_3 and its Fermi-liquid nature, we have performed high-pressure studies on the susceptibility, on the superconducting transition temperature T_c, and on the upper critical field near T_c.

The magnetization studies (performed in Amsterdam) made use of an induction method. A polycrystalline UPt_3 sample (cylinder, $\phi=6$ mm, $l=8$ mm), prepared by arc melting and casting of the material into a water-cooled crucible, was placed in a diamagnetic Cu-Be cell and moved between the centers of two oppositely wound pickup coils. Solid helium served as the pressure transmitting medium. Hydrostatic conditions were preserved as well as possible by freezing the helium under constant pressure conditions. The detection limit of this magnetometer is 5×10^{-6} A m^2. For the UPt_3 sample, the pressure-independent diamagnetic contribution to the induction voltage amounted to 11%, for which a correction was applied. Temperatures were determined from a nearly field-insensitive carbon-glass resistance thermometer. The field effect on the thermometer caused deviations in the temperature of less than 0.2 K in the temperature range 4.2–40 K in a field of 5.3 T; these deviations did not affect the pressure effect on the temperature at which the maximum in χ occurs.

Both T_c (midpoint) and the slope of the upper critical field near T_c, $B'_{c2} \equiv -\partial B_{c2}/\partial T|_{T_c}$, were determined resistively by a standard four-terminal ac technique with the current direction along the hexagonal c axis. These measurements were performed at Los Alamos on a twinned crystal of UPt_3 that was grown in a bismuth flux and subsequently annealed at 1200 °C for 40 h and 1100 °C for an additional 20 h. Pressures up to 19 kbar were generated in a Cu-Be self-clamping cell, modified slightly from the one described in detail elsewhere.[10] Its principle of operation is identical to our earlier design. The pressure at low temperatures was deduced from the inductively measured T_c of a high-purity tin manometer. Great care was taken to ensure thermal equilibration between the sample and the temperature-sensing thermometer, located outside the high-pressure volume.

Magnetization curves of polycrystalline UPt_3 at 4.2 K are shown in Fig. 1 for two different pressures: 1 bar and 4.5 kbar. The value for the relative pressure derivative of the susceptibility, $\partial \ln\chi/\partial P$, as derived from the data of Fig. 1, is -24 Mbar^{-1}. The zero-pressure susceptibility derived from these data is 104×10^{-9} $\text{m}^3/$(mole formula units) and is larger than the value expected for a polycrystalline sample with random orientation of the crystallites, pointing to preferential orientation in this sample. [The susceptibility in the hexagonal plane is 110×10^{-9} $\text{m}^3/$(mole formula units) at 4.2 K (Ref. 4).]

The temperature dependence of the magnetic moment of UPt_3, in a field of 5.3 T, has been measured between 4.2

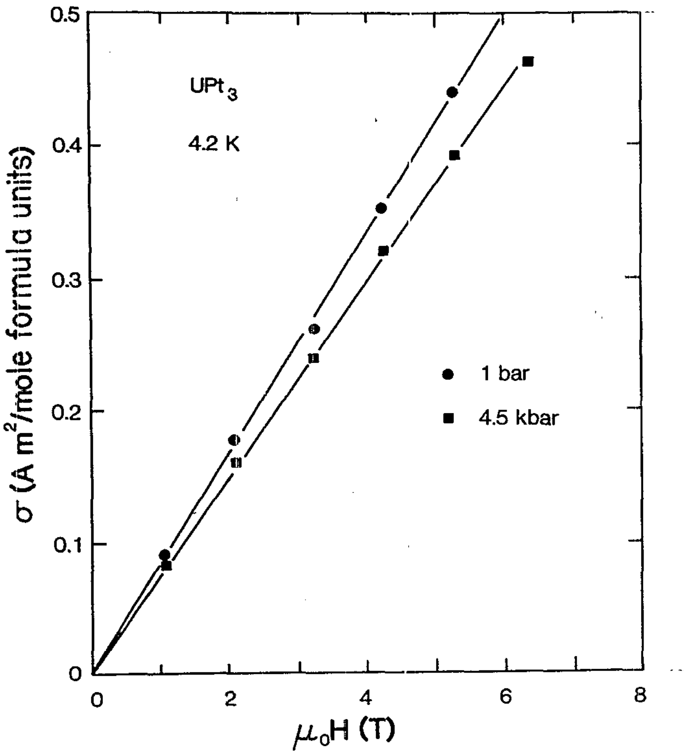

FIG. 1. Magnetization curves for polycrystalline UPt$_3$ at 4.2 K at the pressures indicated.

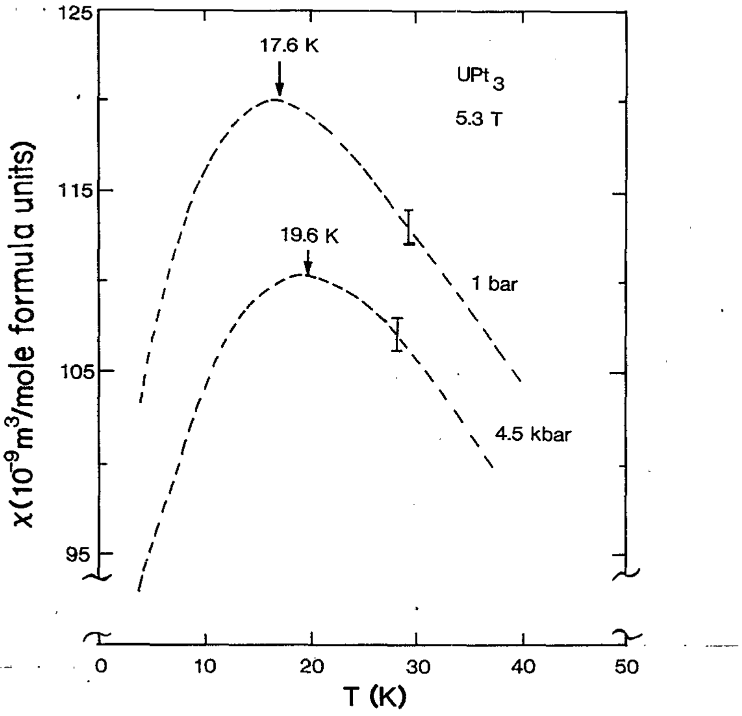

FIG. 2. dc susceptibility as a function of temperature at different pressures for polycrystalline UPt$_3$. The arrows indicate T_m the temperature for which the susceptibility maximum occurs.

and 40 K, at zero pressure and at 4.5 kbar. From these data the temperature dependence of the susceptibility has been determined and, in particular, the temperature T_m where the maximum in the susceptibility occurs and its pressure dependence $\partial T_m/\partial P$ (see Fig. 2). The temperature T_m shifts towards higher temperatures with a rate of 2 K over 4.5 kbar, resulting in a relative pressure dependence $\partial \ln T_m/\partial P$ of 25 Mbar^{-1}, which is almost equal to the value for $\partial \ln \chi/\partial P$. Therefore, the product χT_m may be considered as pressure independent.

The resistivity versus temperature data for UPt$_3$ up to 2 K, plotted on a log-log scale, yield a slope of 2.0 ± 0.1 for each of the applied pressures. Therefore, we display the resistivity versus T^2 in Fig. 3. Clear deviations from T^2 behavior were apparent for temperatures above about 1.5 K, with the resistivity increasing less rapidly than T^2. The (extrapolated) residual resistivity is independent of pressure at the 1% level. The residual resistance ratio, $\rho(300\ \text{K})/\rho(0\ \text{K})$, is approximately 280 at 1 bar, the highest value reported to date for UPt$_3$. We estimate the residual resistivity of this sample to be about $0.5 \times 10^{-8}\ \Omega\,\text{m}$ based on a 300-K c-axis resistivity value of $130 \times 10^{-8}\ \Omega\,\text{m}$.[5]

Figure 3 also shows the systematic decrease of the slope $\partial\rho/\partial T^2$ with pressure. In the spin-fluctuation model of Kaiser and Doniach,[9] the resistivity for $T << T_s$, the spin-fluctuation temperature, is proportional to $(T/T_s)^2$. The determination of T_s is not clear cut for UPt^3, as discussed by Wire *et al.*[8] However, in our analysis, it is only necessary to determine relative changes in T_s and not absolute values. We choose the peak in susceptibility (Fig. 2) as representative of T_s, i.e., $T_s(P=1\ \text{bar}) = 17.6$ K. Therefore, employing $\rho = \rho_0 + A(T/T_s)^2$, we calculate A at 1 bar, and assuming that A is not a function of pressure, we then calculate $T_s(P)$ from the data. The results are shown in Fig. 4. The relative pressure change of T_s, $\partial \ln T_s/\partial P$, is 25 Mbar^{-1}. At low temperatures, a Fermi liquid, such as UPt$_3$, is expected to obey the relation $\chi(T=0) = C/T_s$, where C is an appropriate Curie constant. This implies[8] that $\partial \ln \chi/\partial P = -\partial \ln T_s/\partial P$, so that the effect of pressure on the spin-fluctuation resistivity of UPt$_3$ is $\partial \ln \chi/\partial P = -25$ Mbar^{-1}. This agrees very satisfactorily with the direct measurement of the susceptibility at 1 bar and 4.5 kbar on polycrystalline UPt$_3$ ($\partial \ln \chi/\partial P = -24$ Mbar^{-1}) discussed above. Wire *et al.*[8] report similarly good agreement

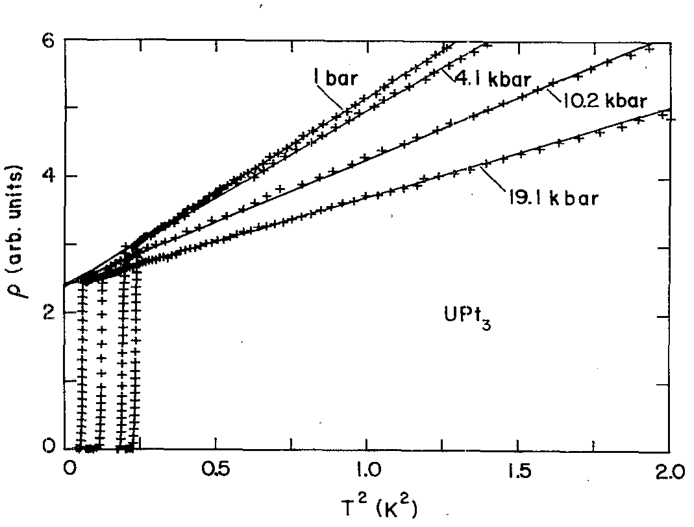

FIG. 3. Resistance vs temperature squared for single-crystal UPt$_3$. Pressures were applied in the order: 1 bar, 10.2 kbar, 19.1 kbar, and 4.1 kbar. The straight lines are a guide to the eye.

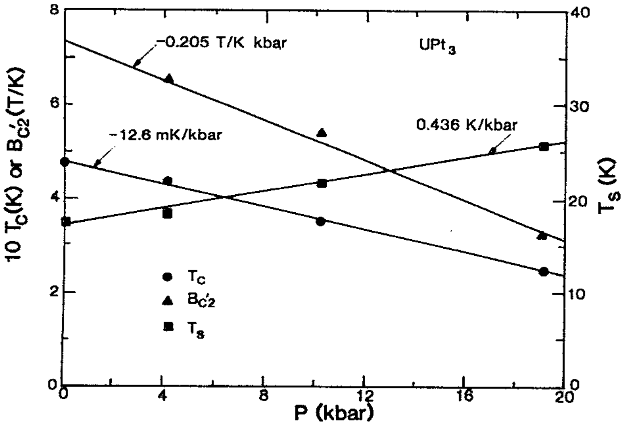

FIG. 4. The spin-fluctuation temperature T_s, the superconducting transition temperature T_c, and the initial slope of the upper critical field $B'_{c2} \equiv -\partial B_{c2}/\partial T|_{T_c}$ vs pressure for single-crystal UPt$_3$.

between χ derived from T_s and χ measured directly for the spin-fluctuation system UAl$_2$ under pressure. These same authors report a somewhat larger value for $\partial \ln\chi/\partial P$ of -30 Mbar^{-1} for UPt$_3$ from c-axis resistivity down to 1 K. We believe that the present value of $-(24$ to $25)$ Mbar^{-1} represents a more accurate result than was previously attainable.

We now discuss the effects of pressure on superconductivity in UPt$_3$. Figure 3 shows the depression of T_c with pressure; the results are replotted in Fig. 4. The relative depression of T_c with pressure $\partial \ln T_c/\partial P$ is -26 Mbar^{-1}. The initial slope of the upper critical field B'_{c2} is also depressed with pressure at a relative rate of -28 Mbar^{-1}. Based on our data to 19 kbar, both T_c and B'_{c2} extrapolate to zero at 37 ± 1 kbar, corresponding to a volume change $\Delta V/V_0$ of -0.018.[11]

We have analyzed the effect of pressure on the electronic specific-heat coefficient γ using our measured values of T_c, B'_{c2}, and ρ and the equations derived from the Bardeen-Cooper-Schrieffer (BCS) theory found in Orlando, NcNiff, Foner, and Beasley.[12] Assuming the dirty limit (mean free path l much shorter than the superconducting coherence length ξ_0) and using the measured γ value $(=1.06\times10^4$ J/m^3 K$^2)^1$, a value for B'_{c2} of 0.34 T is obtained which is only about 5% of the measured value. Clearly, our UPt$_3$ is not in the dirty limit. A knowledge of the Fermi-surface area is required for use of the clean limit or the full expression relating γ to T_c, B'_{c2}, and ρ_0. If we use 1-bar data to calculate the Fermi-surface area and also make the assumption that it is pressure independent, then we may calculate γ as a function of pressure. We find, using either the full or the clean limit formula, that γ is essentially constant to 10 kbar and drops less than 10% at 19 kbar, even though T_c and B'_{c2} have changed by 50%. If our above stated assumptions and the use of these equations are justified, then we find that superconductivity in UPt$_3$ is not as strongly correlated with γ as is the case for high-T_c $A15$ superconductors.[13] [We note that, given our assumptions in calculating $\gamma(P)$, by 19 kbar χ/γ has decreased by about 40% or the Fermi-surface area has decreased by the same amount for χ/γ independent of pressure.]

We note finally that

$$\partial \ln\chi/\partial P \cong \partial \ln T_c/\partial P (\approx \partial \ln B'_{c2}/\partial P) \cong -25 \text{ Mbar}^{-1} \ .$$

This is suggestive of a correlation between the large susceptibility (spin-fluctuation phenomenon) and superconductivity in UPt$_3$. Although a decrease in T_c with pressure is not unusual, Tachiki, Maekawa, and Takahashi[14] suggest that a depression of T_c should occur if the superconductivity is not of the conventional BCS $(S=0)$ type, but rather is of a type enhanced by spin fluctuations. We suggest that this may be the case for UPt$_3$. We note that for other heavy-fermion systems T_c has been observed to increase with pressure in CeCu$_2$Si$_2$ (Ref. 15) and to decrease with pressure in UBe$_{13}$.[16] Furthermore, Valls and Tešanović[17] predict, within a Fermi-liquid model in the limit of large effective electronic mass, that T_c should be proportional to T_s, or inversely proportional to χ, which is the opposite of what we find here for UPt$_3$. We can only comment that the different pressure dependences observed experimentally for these three heavy-fermion systems are as perplexing as are the different theoretical predictions.

In summary, we have performed resistivity and susceptibility measurements on single-crystal and polycrystalline UPt$_3$ under pressures up to 19 kbar. Superconductivity and spin fluctuations are both found to be strongly depressed by pressure with

$$\partial \ln T_c/\partial P \cong \partial \ln\chi/\partial P = -25 \text{ Mbar}^{-1} \ .$$

The correlations between T_c and $\chi(0)$ are highly suggestive of a non-BCS-type pairing mechanism.

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