Observation of High-Field Superconductivity of a Strongly Interacting Fermi Liquid in U_6Fe

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Measurements of the temperature dependences of the upper critical field H_{c2} and heat capacity C_P of U_6Fe are reported. H_{c2} increases very rapidly for a low-transition-temperature ($T_c = 3.8$ K) superconductor, reaching $H_{c2} = 64$ kOe at $T = 1.95$ K. C_P data indicate that U_6Fe is a strong-coupled bulk superconductor and an exchange-enhanced paramagnet with an electronic coefficient $\gamma = 145 \pm 10$ mJ/mole·K^2. U_6Fe constitutes a metallurgically clean and conclusive example of the occurrence of high-field superconductivity in a strongly interacting Fermi liquid.

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Superconductivity and magnetism are generally acknowledged to be mutually exclusive phenomena. In spite of this trend, superconductivity has recently been detected in two strongly magnetic materials: $CeCu_2Si_2$ (Ref. 1) and Y_9Co_7 ("Y_4Co_3").[2] Unfortunately, metallurgical difficulties have so far prevented clear interpretations of these experiments.[3]

We report our observations of high-field superconductivity and exchange-enhanced paramagnetism in the compound U_6Fe, and present conclusive evidence that these phenomena occur within a strongly interacting Fermi liquid. U_6Fe is presently unique in that it is clearly a metallurgically clean, bulk superconductor with very reproducible properties, and does not exhibit localized magnetic moment behavior as does the "Kondo lattice," $CeCu_2Si_2$.[3-5] Further, we find that the renormalization of the Fermi-liquid parameters of U_6Fe is much stronger, and the $T_c = 3.8$ K much smaller, than appropriate to high-field transition-metal compounds.

Only a handful of superconducting binary U compounds (UCo and U_6X, X = Mn, Fe, Co, Ni) are presently known.[6,7] These materials are among the few superconducting compounds of the magnetic $3d$ elements Mn through Ni, and recent measurements[5] have shown that U_6Fe and U_6Co have strong, weakly temperature-dependent paramagnetic susceptibilities comparable with that of the nonsuperconducting, nearly ferromagnetic element, Pd. The body-centered tetragonal ($D2_c$) crystal structure[8] of U_6X is unique to U, Np, and Pu compounds with magnetic $3d$ elements.[9] These intriguing observations suggest that high-mass, itinerant $5f$ electrons play a significant role in the physical properties of these materials.

An 11-g ingot of composition $U_{6.1}Fe$ was prepared by the arc melting of high-purity starting materials in an Ar atmosphere followed by an annealing procedure which is described elsewhere.[10] Samples were spark cut from the annealed ingot for C_P and H_{c2} measurements and another portion of the original material was analyzed by x-ray diffraction and found to be single phase. The temperature dependence of the upper critical field was determined resitively by varying of the sample temperature at fixed values of field H. No transition hysteresis was observed when temperature was cycled through $T_c(H)$. Specific heat was measured by use of a standard semiadiabatic, heat-pulse method.

Our results for H_{c2} vs T are given in Fig. 1. H_{c2} increases linearly with decreasing temperature over the entire experimental range $2 \lesssim T \lesssim 4$ K and $0 \leq H \lesssim 65$ kOe. *The magnitude of* $H_{c2}(T \to 0)$ *approaches 100 kOe, a value which is anomalously large for a compound with* $T_c \lesssim 4$ *K.* The slope $-(dH_{c2}/dT)_{T_c} = 34.2$ kOe/K rivals similar data for the extreme high-field $A15$ and Chevrel-phase superconductors for which

$$20 \text{ kOe/K} \lesssim -(dH_{c2}/dT)_{T_c} \lesssim 80 \text{ kOe/K}$$

and $T_c > 10$ K.

Zero-field data for C_P vs T were obtained over the range $1 \lesssim T \lesssim 20$ K, and our results for C_P/T vs T^2 are shown in Fig. 2(a). We note that the low-temperature C_P of U_6Fe is roughly one order of magnitude larger than that of typical transition-metal compounds (including $A15$'s) over a similar temperature range. C_P data obtained in an applied field $H = 2$ kOe yielded $-(dH_{c2}/dT)_{T_c} = 36.4$ kOe/K, in good agreement with the H_{c2} measurements.

The unusual negative curvature of C_P/T vs T^2

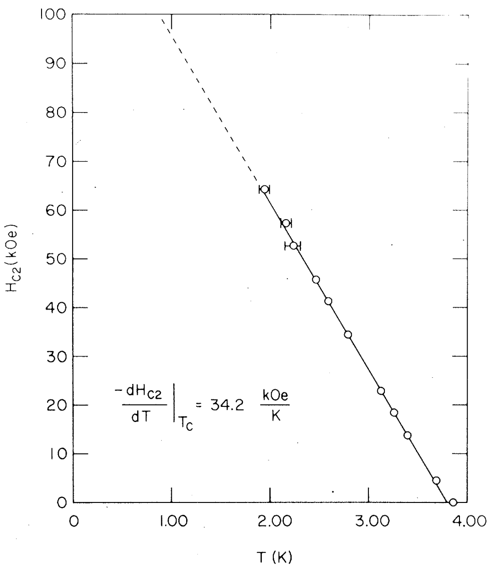

FIG. 1. Upper critical magnetic field H_{c2} vs temperature T for U_6Fe. The line is a guide to the eye, and has the slope shown.

for $T > T_c = 3.70$ K has been observed previously for the highest-T_c $A15$ (Refs. 11 and 12) and Chevrel-phase[13] compounds where it has been attributed to low-energy features in the phonon density of states. A plot of C_P/T vs T shown in Fig. 2(b) further illustrates this remarkable behavior.

We have fitted our normal-state data over limited temperature ranges using either a modified Debye expression

$$C_{n1} = \gamma_1 T + \beta_1 T^3 + \alpha_1 T^5, \qquad (1)$$

or an alternative expression

$$C_{n2} = \gamma_2 T + \delta_2 T^2 + \beta_2 T^3, \qquad (2)$$

which reflects the dominant T^2 behavior of C_P vs T shown in Fig. 2(b) (see Table I). We are unable to fit our entire data set for the normal-state heat capacity C_n by a low-power polynomial in T and retain consistency with entropy constraints which require $\gamma > 100$ mJ/mole$\cdot$K^2. The coefficients of fit No. 1 are in very good agreement with the unpublished results of Maita,[14] and our large γ value is corroborated by two additional observations.

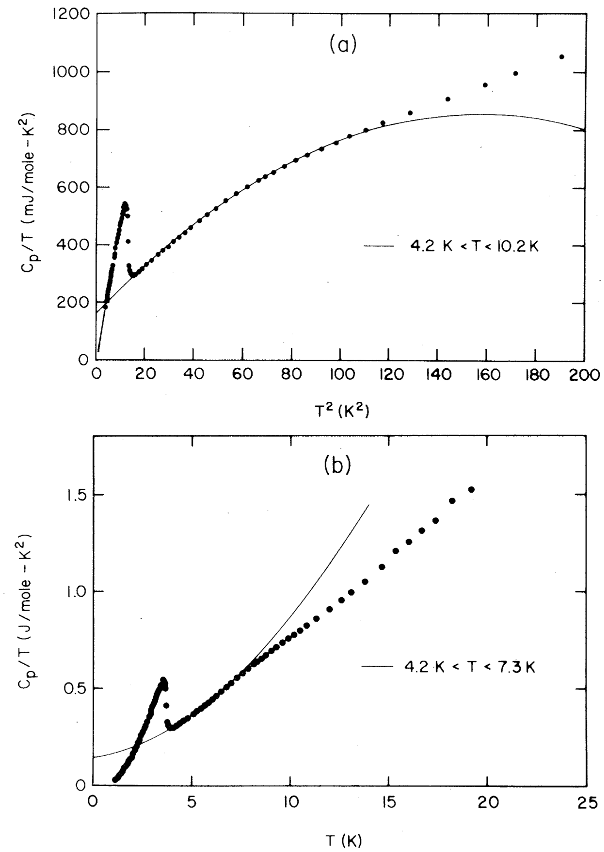

FIG. 2. (a) Zero applied magnetic field heat capacity C_P divided by temperature T vs T^2 for U_6Fe. The thick solid line represents data which were too dense to plot, and the thin solid line represents the fit No. 1 of data from $4.2 \leq T \leq 10.2$ K, as described in the text. Note the superconducting transition anomaly at $T_c = 3.70$ K. (b) Zero applied magnetic field C_P/T vs T for the same U_6Fe data shown in (a) above. The thin solid line represents the fit No. 2 data from $4.2 \leq T \leq 7.3$ K, as described in the text.

(1) The BCS theory predicts the magnitude of

TABLE I. Fitting parameters for the normal-state specific heat of U_6Fe.

		γ (mJ/mole$\cdot$K^2)	δ (mJ/mole$\cdot$K^3)	β (mJ/mole$\cdot$K^4)	α (mJ/mole$\cdot$K^6)	θ_D [a] (K)	$S_n(T_c)/S_s(T_c)$ [b]
Fit No. 1	$4.2 \leq T \leq 10.2$ K	155.2	...	8.954	-2.907×10^{-2}	115	1.00
Fit No. 2	$4.2 \leq T \leq 7.2$ K	136.9	17.88	5.372	...	136	1.00

[a] Debye temperature dependence deduced from β coefficient.
[b] Ratio of the normal to superconducting state entropies at T_c (thermodynamics demands that this ratio equal 1.00).

the jump ΔC in heat capacity at T_c:

$$\Delta C \equiv C_n - C_s = (1.43)\gamma T_c. \quad (3)$$

We estimate $\Delta C = 1.2$ J/mole·K, implying $\gamma \lesssim 230$ mJ/mole·K^2, according to Eq. (3). The actual γ may be somewhat smaller because of strong-coupling effects; our $\gamma_1 = 155$ mJ/mole·K^2 leads to $\Delta C/\gamma_1 T_c = 2.1$, a value typical of a strong-coupled superconductor.

(2) $(dH_{c2}/dT)_{T_c}$ has been successfully correlated[15] with γ and the electrical resistivity ρ of Chevrel-phase compounds by use of the dirty-limit formula

$$(dH_{c2}/dT)_{T_c} = (44.4)\gamma\rho. \quad (4)$$

Using our results

$$-dH_{c2}/dT = 34.2 \text{ kOe/K},$$

$$\gamma_1 = 1.855 \times 10^4 \text{ ergs/cm}^3\cdot\text{K}^2,$$

we deduce $\rho \approx 4.2 \times 10^{-5}$ Ω cm in very good agreement with our measured $\rho(T = 4 \text{ K}) = 5.0 \times 10^{-5}$ Ω cm and other published results for U_6Fe.[16]

The above two observations confirm that *the superconductivity of U_6Fe is a bulk phenomenon which occurs within a strongly interacting Fermi liquid.* Indeed, our $\gamma \approx 155$ mJ/mole·K^2 for U_6Fe is comparable with values of 142 and 171 mJ/mole·K^2 reported for the nonsuperconducting spin-fluctuation compounds, UAl_2 (Ref. 17) and USn_3,[18] respectively. The magnitude of the low-temperature magnetic susceptibility[5]

$$\chi \approx 2 \times 10^{-6} \text{ cm}^3/\text{g} \approx 4 \times 10^{-4} \text{ cm}^3/\text{mole}\cdot\text{atom}$$

for U_6Fe is approximately five times less than the more temperature-dependent susceptibilities of UAl_2 (Ref. 17) and USn_3,[19] and about one-half as large as that of Pd.[20] It is therefore of interest to estimate the degree of exchange enhancement of the electronic spin susceptibility $\chi_s = \chi_P \mathcal{S}$ ($\chi_P \equiv$ Pauli spin susceptibility) for U_6Fe.

$\mathcal{S}$ can be derived from the relation

$$(1+\lambda)\mathcal{S}^{-1} = 3\left(\frac{\mu_B}{\pi k_B}\right)^2 \frac{\gamma}{\chi_s} = \frac{N_\gamma}{N_\chi}, \quad (5)$$

where N_γ and N_χ are the renormalized densities of states determined from specific-heat and susceptibility measurements, respectively, μ_B is the Bohr magneton, k_B is Boltzmann's constant, and λ is the electron-phonon interaction parameter obtained from the McMillan equation for T_c.[21] Using $\chi = 2.0 \times 10^{-6}$ cm^3/g and performing standard corrections for core and band diamagnetism, we obtain $\mathcal{S} \approx 4$, indicating that magnetic correlations are significant in U_6Fe.

In view of the apparent strong paramagnetism of U_6Fe, we have also analyzed our data within a paramagnon model[22,23] in which $\lambda \to \lambda + \lambda_s$ in Eq. (5) and λ_s is a paramagnon interaction parameter. We replace the McMillan formula by[24]

$$T_c = \frac{\omega_c}{1.2} \exp\left(\frac{(1+\lambda+\lambda_s)}{(\mu^* + \lambda_s - 1)}\right). \quad (6)$$

Using a Coulomb interaction parameter $\mu^* = 0.13$ and the experimental ratio $N_\gamma/N_\chi \approx 0.5$, we conclude that $5 \lesssim \mathcal{S} \lesssim 10$ for $0.4 \lesssim \lambda_s \lesssim 1.5$, corresponding to $0.9 \lesssim \lambda \lesssim 2.1$ (these estimates are consistent with a wide range of characteristic phonon energy, $40 \lesssim \omega_c \lesssim 240$ K).

Strong renormalizations of the electronic heat capacity and magnetic susceptibility have important implications for the critical-field behavior of U_6Fe. Orlando and Beasley[24] have shown that H_{c2} data of $A15$ compounds are best understood by taking into account the full renormalization of the paramagnetic limiting field H_P and the effects of paramagnon suppression of superconductivity without invoking unreasonably large spin-orbit scattering rates. Accordingly, we assume a second-order transition in the dirty limit where

$$H_P(0) = H_P^{\text{BCS}}(0)(1+\lambda+\lambda_s)/\mathcal{S}$$
$$= (18.6 \text{ kOe/K}) T_c N_\gamma/N_\chi. \quad (7)$$

Our data yield $H_P(0) \approx 34$ kOe $\ll 64$ kOe, the largest value of H_{c2} observed in our experiments, implying that the inclusion of a large amount of spin-orbit scattering may yet be necessary to explain our results. Such a possibility is of importance in view of existing difficulties in theories of H_{c2}.[24,25]

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Note added.—After submission of this paper for publication, we have become aware of similar results of H. R. Ott *et al.* for the compound UBe_{13}.[26] Although UBe_{13} exhibits Curie-Weiss behavior for χ at $T \gtrsim 100$ K, the scaling relations of Eqs. (3) and (4) above are still satisfied, suggesting that UBe_{13} is an even more strongly interacting Fermi liquid with a bandwidth roughly one-tenth that of U_6Fe. Our results and those of Ott *et al.*

demonstrate that a model of "heavy fermion" superconductivity can be applied to even nearly ferromagnetic materials, and over at least a three-order-of-magnitude range of bandwidth.

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