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HIGH-FIELD, HIGH- T_c SUPERCONDUCTIVITY IN U_6Fe AND U_6Co *

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HIGH-FIELD, HIGH- T_c SUPERCONDUCTIVITY IN U_6Fe AND U_6Co *

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Upper critical field data for U_6Fe and U_6Co are found to be markedly inconsistent with the weak coupling WHHM model. Nevertheless, excellent quantitative fits of H_{c2} vs T are obtained by using a modification of the WHHM model in which the orbital field is multiplied by a temperature-dependent enhancement factor.

Keywords: superconductivity, actinides, type-II superconductivity.

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The U_6X ($X=Mn, Fe, Co, Ni$) compounds are under active investigation for several reasons. Their physical properties, as well as recent phenomenological arguments, have suggested that these superconductors are near a magnetic instability [1,2]. These materials possess unusually large electronic effective masses $m^* \lesssim 25 m_e$ ($m_e \equiv$ free electron mass), and provide an important link between narrow-band transition metals and heavy fermion superconductors such as UPt_3 , UBe_{13} , and $CeCu_2Si_2$. Indeed, high- T_c , high-critical field A15 and Chevrel compounds have many properties in common with U_6Fe , the highest- T_c (≥ 4 K), highest- H_{c2} (≤ 12 T) U compound presently known (UBe_{13} also has $H_{c2}(T=0) \sim 10-12$ T [3,4]). We have observed particularly interesting behavior of $H_{c2}(T)$ in U_6Fe and U_6Co samples, as we discuss below.

We have performed resistive and inductive measurements of $H_{c2}(T)$ on polycrystalline samples of various purities by sweeping both field and temperature. Representative resistive data are illustrated in Fig. 1, which gives our results for the highest purity U_6Fe sample (II47B) that we have examined. These $H_{c2}(T)$ data exhibit clear positive curvature to fields in excess of 3 T, followed by a quasi-linear behavior to the remarkably high $H_{c2} \sim 10$ T at the lowest temperatures that we have attained. These characteristics cannot be understood within the traditional dirty limit WHHM model [5], as shown in Fig. 1. We emphasize that reasonable adjustments of the initial slope $H'_{c2}(T_{co})$ or the introduction of artificially high values of the spin-orbit scattering parameter λ_{so} do not yield appreciably better agreement between the model and our experimental results.

Measurements on lower purity U_6Fe samples are illustrated in Fig. 2. These data show that the positive curvature of the phase boundary decreases with decreasing residual resistivity ratio RRR, and reveal evolutions of $T_{co}(H=0)$, $H'_{c2}(T_{co})$ and $H_{c2}(T=0)$ similar to those observed in A15 and Chevrel compounds of variable quality (see Table 1) [6,7]. We doubt that the positive curvature

Fig. 1

Greek
lamda

Fig. 2

Table 1

observed at moderate field strengths is due to sample inhomogeneities [7], since small amounts of more pronounced positive curvature ("tails") are observed in even lower purity samples (such as II51) at much lower fields $\lesssim 0.2$ T. We do consider these latter features to be spurious and have ignored them in our analyses. We note that estimates of the zero temperature coherence length ξ_0 and the transport mean-free path l_{tr} (see Table 1) show that all samples are in the dirty limit of WHHM.

Orlando and coworkers [6] have found it necessary to introduce many-body renormalizations of the Pauli susceptibility and m^* in order to explain anomalous paramagnetic limiting in the higher-field H_{c2} data for several A15 compounds. Nevertheless, Decroux and Fischer [7] were unable to use similar arguments to account for the high H_{c2} values that they observed for $Mo_6(Se_{1-x}S_x)_8$ materials, whose normal and superconducting state properties are somewhat similar to the U_6X compounds. The central difficulty is that the WHHM model cannot describe materials for which $H_{c2} > H_{c2}^*$ where H_{c2}^* is the orbital upper critical field attained in the absence of paramagnetic limiting (i.e., the case $\lambda_{s0} \rightarrow \infty$).

Decroux and Fischer were forced to introduce a phenomenological function $\beta(T)$ which multiplied the orbital field according to

$$H_{c2}^{**}(T) \equiv \beta(T) H_{c2}^*(T) = \frac{\beta_0 H_{c2}^*(T)}{1 + (\beta_0 - 1) (T/T_{c0})^2} \quad (1)$$

Substitution of the resulting enhanced orbital field H_{c2}^{**} brought the WHHM model into agreement with their Chevrel compound data (using only physically realistic values of λ_{s0}).

Noting similarities in the temperature dependences of the heat capacity, electrical resistivity and H_{c2} between $Mo_6(Se_{1-x}S_x)_8$ and U_6X materials [1,7,8],

we have applied Eq. 1 to our U_6Fe and U_6Co results. Figure 1 is illustrative of the remarkably good agreement obtained between the modified WHHM model and experiment.

We must point out that the actual application of Eq. 1 in fits of the data is, in fact, restricted to replacing $H_{c2}^*(0)$ by $\beta(T)H_{c2}^*(0)$ wherever it occurs in orbital terms of the WHHM equation for $H_{c2}(T)$ [7]. We have also adopted this empirical rule and, in this narrow sense, our procedure is evidently identical to that used by Decroux and Fischer in their Chevrel compound analyses. However, the success of the fitting procedure does not necessarily imply the general validity of Eq. 1 at arbitrary temperatures. We have performed point-by-point, force-fits of the H_{c2} data in order to compare our best-fit $\beta(T)$ (assuming the form of Eq. 1) to those values of $\beta(T)$ necessary for obtaining exact correspondence between the WHHM model (using $H_{c2}^*(0) + \beta(T) H_{c2}^*(0)$) and experiment. The point-by-point values of $\beta(T)$ were then fitted to the function given in Eq. 1, and the results are shown in the inset to Fig. 1. It is clear that the functional form of β given in Eq. 1 is an excellent approximation for the empirical $\beta(T)$ function necessary to reproduce the observed phase boundary over the entire experimental range.

We have also analyzed the effects of a many-body renormalization of the fermion mass and the exchange enhancement S of the Pauli susceptibility on the Maki parameter α^* and the Pauli limiting field $H_p^*(0)$ using the following relations:

$$\alpha^* = \frac{\sqrt{2} H_{c2}^{**}(0)}{\beta_0 H_p^*(0)} = (-0.528 \text{ K/T}) \frac{S H_{c2}^*(T_{c0})}{m^*/m_e} \quad (2)$$

$$H_{c2}^{**}(0) = (-0.693) \beta_0 T_{c0} H_{c2}^*(T_{c0}) \quad (3)$$

Greek
alpha

$$H_p^*(0) = (1.86 \text{ T/K}) \frac{T_{co} m^*/m_e}{S} \quad (4)$$

$$S = \frac{m^*/m_e}{1 + F_0^a} \quad (5)$$

$e_l=0$ F_0^a is the antisymmetric, $l=0$ Landau parameter, and we set $m^* \sim 20 m_e$ for U_6Fe and U_6Co for illustrative purposes. We have also fixed $\lambda_{so} = (2/3) \lambda_{tr}$, which is the maximum value that is consistent with the measured electrical resistivity, in order to minimize the magnitude of β_0 . A summary of parameters obtained from fits of the data for several samples is given in Table 2.

Table 2

The fits are somewhat insensitive to the exact values of β_0 and α^* --that is, a larger value of β_0 can be balanced by a larger value of α^* , holding λ_{so} fixed. This situation is highlighted in Table 2, where we have given ranges of parameters that provide good fits of the data. Future studies must therefore seek to clarify the physical significance and magnitude of $\beta(T)$ in order to deduce reasonable estimates of α^* , S and F_0^a from such fits. We are nevertheless able to observe that U_6Co is more Pauli limited than U_6Fe , and that the purer samples require larger enhancements of H_{c2}^* and are less Pauli limited than the dirtier materials.

Decroux and Fischer have proposed several origins for the enhancement function $\beta(T)$ [7]: 1) Strong coupling corrections can lead to significant increases in H_{c2}^* at lower temperatures when there is a large strength of the electron-phonon spectral function $\alpha^2(\omega)F(\omega)$ at low frequency. 2) Anisotropy effects (including nonlocal corrections to the gap equation, anisotropy of the electron-phonon interaction and band structure anisotropy) can cause positive curvature of the phase boundary; however, these effects are expected to decrease rapidly with decreasing sample purity. 3) The presence of different bands

Greek
omega

having significantly different Fermi velocities and scattering lifetimes may account for the observed behavior. Decroux has shown [7] that when the ratio of Fermi velocities is 1:3 in a model two-band material and the interband scattering is weak, a phase boundary similar to that of Fig. 1 is predicted.

We believe that many of the similarities between Chevrel phase and U_6X materials are likely to be due to the presence of low frequency "cluster modes" [9] in the phonon density of states of both types of materials. Evidence for cluster modes in U_6X compounds can be found in recent heat capacity [8,11] and neutron diffraction [10] experiments. Although all of the mechanisms 1) through 3) above could easily contribute to the anomalous H_{c2} behavior of U_6Fe and U_6Co , the large m^* and 5f bonding characteristic of these materials leads us to consider additional mechanisms. More extensive experiments on single crystals and other compounds of variable purity are currently underway to clarify the physical origin of $\beta(T)$.

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Figure Captions

Fig. 1. Upper critical field H_{c2} versus temperature T for U_6Fe sample II47B, whose zero-field transition temperature $T_{co} = 3.98$ K and residual resistance ratio (at 4.2 K) $RRR = 9.6$. The dashed line represents the prediction of the WHHM model with $\lambda_{so} = 5.0$ and $H'_{so}(T_{co}) = 2.83$ T/K. The solid line represents the modified WHHM model fit with $\lambda_{so} = 5$, $\beta_0 = 1.41$, and $T_{co} = 3.94$ K, as described in Table II. The solid points are experimental data. Inset: Empirical values of the orbital field enhancement function $\beta(T)$ versus T for U_6Fe sample II47B. The points represent values of $\beta(T)$ required by a point-by-point force-fit of the data, and the solid line is a best-fit of these points to the form implied by Eq. 1 of the text, yielding $\beta_0 = 1.41$ and $T_{co} = 3.94$ K, in excellent agreement with the modified WHHM fit of $H_{c2}(T)$.

Fig. 2. Upper critical field H_{c2} versus temperature T for three U_6Fe samples of different purity (see Table 1 for sample properties). The lines represent smooth fits of the experimental data using the modified WHHM model of Eq. 1. The values of the residual resistance ratio RRR for the samples are: 9.6 (II47B), 2.0 (II33), and 1.6 (II51).

Table 1. Experimental Parameters of Samples Studied

Sample	RRR (at 4K)	$T_{CO}^{\dagger\dagger}$ (K)	γ^{*+} (J/m^3K^2)	$H_{c2}^i(T_{CO})^{\dagger\dagger}$ (T/K)	$\rho(T_{CO})$ ($\Omega \cdot m$)	$\epsilon_0^{\dagger\dagger}$ (m)	$l_{tr}/\epsilon_0^{\dagger\dagger}$
U ₆ Fe (II47B)	9.6	3.98(3.92)	1.76×10^3	-2.83	1.4×10^{-7}	3.14×10^{-8}	1.09×10^{-1}
U ₆ Fe (II33)	2.0	3.85(3.81)	1.85×10^3	-3.35	5.9×10^{-7}	3.25×10^{-8}	2.57×10^{-2}
U ₆ Fe (II51)	1.6	3.75(3.54)	$1.85 \times 10^{3\dagger}$	-3.66	1.1×10^{-6}	3.33×10^{-8}	1.40×10^{-2}
U ₆ Co (II53B)	3.6	2.56(2.53)	$1.60 \times 10^{3\dagger}$	-3.27	6.1×10^{-7}	4.88×10^{-8}	1.67×10^{-2}
U ₆ Co (529)	1.3	2.54(2.45)	$1.60 \times 10^{3\dagger}$	-3.89	--	$4.88 \times 10^{-8\dagger}$	--

[†]Estimated from data on other samples.

^{††}Experimental zero field values are given. T_{CO} values used in the unmodified WHHM fits are given in parentheses. The experimental $H_{c2}^i(T_{CO})$ values were used in the unmodified WHHM fits.

[†]After refs. 8 and 11.

^{††}Estimated using a fermion density of 3 per U atom.

Table 2. Fitting Parameters for $H_{c2}(T)$

Sample	T_{co}^\dagger (K)	$H_{c2}^\dagger(T_{co})^\dagger$ (T/K)	λ_{so}	$\beta(0)$	$H_{c2}^{**}(0)$ (T)	α^*	$H_p^*(0)$ (T)	S	F_o^a
U_6Fe (II47B)	3.94	-2.56	5.0	1.41- 1.51	9.83- 10.4	6.29×10^{-2} - 1.16	157- 8.54	0.931- 17.2	20.5- 0.163
U_6Fe (II33)	3.84	-2.98	20.0	1.37- 1.40	10.9- 11.0	7.85×10^{-2} - 1.02	143- 11.0	0.993- 13.0	19.0- 0.538
U_6Fe (II51)	3.54	-3.58	40.0	1.19- 1.26	10.6- 11.0	2.02- 3.47	6.15- 3.59	21.4- 36.7	-0.065 to -0.455
U_6Co (II53B)	2.52	-3.22	36.0	1.17- 1.57	6.57- 8.41	2.30- 7.87	3.46- 1.01	27.1- 92.6	-0.262 to -0.784
U_6Co (529)	2.45	-3.88	75.0 ⁺	1.11- 1.54	7.52- 10.1	3.33- 12.7	2.80- 0.735	32.5- 124	-0.385 to -0.839

[†]Insignificant variations of T_{co} and $H_{c2}^\dagger(T_{co})^\dagger$ generally accompanied the larger variations of other parameters obtained in different fits of the data.

⁺Estimated from data on other samples.



