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Shape of the upper-critical-field curves in URu_2Si_2: Evidence for anisotropic pairing

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Measurements of the upper critical magnetic field H_{c2} of single-crystal URu_2Si_2 are reported for fields $H \leq 8$ T and temperatures $T \geq 0.5$ K. H_{c2} is strongly anisotropic and exhibits pronounced positive curvature for H||a in the tetragonal basal plane. These features can be explained using a theoretical model that assumes a multicomponent superconducting order parameter that is strongly coupled to the antiferromagnetic moment.

Heavy-fermion systems display a rich variety of phase transitions to ordered states at low temperatures.[1] Perhaps the most remarkable and most complex of these systems are UPt_3 and URu_2Si_2. With decreasing temperature they first exhibit antiferromagnetism[2-5] and, at lower temperature, superconductivity. Coexistence of superconductivity and magnetic order has been known for some time, notably in ternary rare-earth compounds.[6] In contrast to the latter materials, the relatively strong hybridization between U $5f$ and itinerant band states present in URu_2Si_2 and UPt_3 ensures that the superconductivity and magnetic phase transitions occur within a common set of strongly interacting electronic states near the Fermi energy. Further, the coupling between the two types of order parameter is expected to be strong and anisotropic, giving rise to interesting new phenomena. This has been suggested in UPt_3, where a double jump in the heat capacity has recently been observed.[7,8] This is ascribed to a splitting of the critical temperature of the two components[9] of the superconducting order parameter which couple differently to the antiferromagnetism.[10]

In this paper, we measure and present a theory for a different property, the upper critical field, in URu_2Si_2. URu_2Si_2 and UPt_3 differ in the way their respective antiferromagnetic moments break the crystal symmetry, and in the size of their effective masses. Our proposal implies that anisotropic pairing occurs in relatively low-mass ($m^* \sim 25m_e$) materials like URu_2Si_2 (Refs. 4 and 5) as well as high-mass ($m^* \sim 200m_e$) materials like UPt_3.[1]

The samples were grown by vertical float-zone refining. Rods of URu_2Si_2 5 mm in diameter by 40 mm in length were initially cast in an arc-melting apparatus. Two of the rods were then vertically positioned in the zone melter with each rod held at one end by a Ta collet. The zone was formed by a single-turn induction coil operated at about 6 MHz with Ti-gettered Ar flowing over the rods. The two rods were initially fused together to form one long sample 80 mm in length. A zone was then passed through the rod at about 1 cm/h to form the single crystal. The zone traveled about 7 mm before the material became single grained with the resulting crystal having an arbitrary orientation with respect to the rod axis.

Two samples were cut from the crystal into parallelepipeds with dimensions $4.4 \times 0.58 \times 0.95$ and $6.7 \times 0.58 \times 0.64$ mm^3 and mounted such that the magnetic field was parallel to the **c** and **a** axes, respectively. ac transverse magnetoresistance was measured at 100 Hz with the standard four-probe method in fields up to 8 T and temperatures down to 0.5 K in a ^{3}He cyrostat. Electrical leads were attached to the sample with silver paint. The data were taken by two methods, by sweeping the field up to 8 T at several fixed temperatures, and by temperature scans in various fixed fields. Data obtained by both methods were in good agreement.

Figure 1 shows the resistivity versus temperature curve in zero field for one of the samples. The antiferromagnetic peak occurs at $T_N = 16.8$ K followed by a superconducting transition with the midpoint at 1.44 K and $\Delta T_c[(10\text{-}90)\%] = 80$ mK. The second sample showed a slightly lower T_c of 1.41 K and $\Delta T_c(10\text{-}90\%) = 130$ mK. The quality of these crystals is demonstrated by their high transition temperatures which are considerably larger than those reported earlier for single crystals[3] which gave $0.8 \leq T_c \leq 1.16$ K. Likewise, the normal-state resistivity

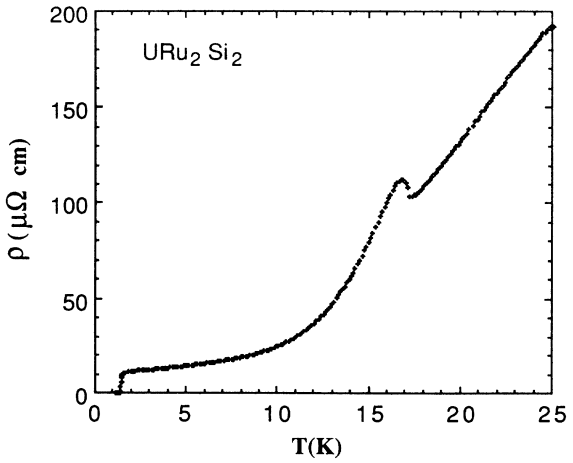

FIG. 1. Resistance vs temperature curve in zero field for the URu_2Si_2 single crystal.

of 11 $\mu\Omega$ cm for the first sample and 14 $\mu\Omega$ cm for the second sample near T_c is about a factor of 3 smaller than reported on the earlier single crystals.[3] The difference in quality between the two crystals as indicated by T_c, ΔT_c, and the normal-state resistivity is probably due to their slightly different locations in the zone refined rod. The residual resistivity ratio $R(300\ \mathrm{K})/R(1.5\ \mathrm{K})$ of heavy-fermion systems is generally not as good a measure of sample quality as in ordinary metals because of the unusual shape of the normal-state resistivity curve. Nevertheless the values of our crystals, ~ 34, is considerably higher than the value of ~ 10 found for earlier crystals.[3]

Magnetoresistance data for the $T_c = 1.44$ K crystal obtained from temperature sweeps at several fixed fields parallel to **a** are shown in Fig. 2. There is a strong, positive normal-state magnetoresistance, as observed previously.[3] From the definition of T_c as the midpoint of these transitions, we construct the upper critical field H_{c2} versus temperature curves for URu_2Si_2 with the field along the **a** and **c** directions, as shown in Fig. 3. Other definitions of T_c such as the 10% or 90% points of the transition give an identical shape with an offset arising from the finite width of the transition curve. Once a definition of T_c is chosen, the smoothness of the data in Fig. 2 leads to an error bar in H_{c2} which is less than the size of the symbols in Fig. 3. A strong anisotropy is observed between the two field directions, in good agreement with earlier low-field measurements.[3] In addition, we observe pronounced upward curvature for the field parallel to the **a** axis which is absent for the field along the tetragonal **c** axis. We attribute this to the existence of a multicomponent superconducting order parameter which couples strongly to the antiferromagnetic order parameter.

Let us assume that URu_2Si_2 is a d-wave superconductor characterized by an order parameter $\Psi = (\Psi_x, \Psi_y)$ which transforms according to the E_{1g} representation of the tetragonal point group. Other two-dimensional representations, including those of p-wave symmetry, can be considered and lead to similar results.[11] Ψ couples to the staggered magnetization $\mathbf{M}_s = \pm M_s \hat{\mathbf{z}}$, where $\hat{\mathbf{z}}$ is along the **c** axis.[3] In zero field, the terms of the Ginzburg-Landau free energy quadratic in Ψ are

$$F_2 = a_0(T - T_{c0})\Psi \cdot \Psi^* - ib\mathbf{M}_s \cdot \Psi \times \Psi^* ,$$

where T_{c0} is the critical temperature if there were no coupling between antiferromagnetism and superconductivity ($b = 0$). In a single magnetic domain, we may take $\mathbf{M}_s = M_s \hat{\mathbf{z}}$. We may also assume that M_s is roughly independent of temperature and field near $T_{c0} \ll T_N$. Defining $\Psi_\pm = (\Psi_x \pm \Psi_y)/\sqrt{2}$, F_2 may be rewritten as

$$F_2 = u_+ |\Psi_+|^2 + u_- |\Psi_-|^2 ,$$

where

$$u_\pm = a_0(T - T_{c0}) \pm bM_s .$$

The actual superconducting transition occurs at $T_c = T_{c0} + |bM_s/a_0|$. $\Psi_\pm$ correspond to Cooper-pair wave functions with different z components of their orbital angular momentum. There is an energy associated with the relative orientation of $\mathbf{M}_s$ and this angular momentum. A second transition may or may not be observed below T_c; this depends on whether or not the fourth-order terms stabilize the phase which occurs precisely at T_c.[12] In UPt_3, such a phase transition to a different low-temperature state is observed.

Now consider the system in an applied field $\mathbf{H} = H\hat{\mathbf{x}}$ along the tetragonal **a** axis. There are gradient terms in the free energy[11]

$$F_g = K_1(|p_y\Psi_x|^2 + |p_y\Psi_y|^2) + K|p_y\Psi_y|^2 + K_4(|p_z\Psi_x|^2 + |p_z\Psi_y|^2) .$$

Here $p_y = -i\partial/\partial y$ and $p_z = -i\partial/\partial z + 2eHy/\hbar c$. We wish to minimize $F = F_2 + F_g$. The first step is to choose Ψ_x and Ψ_y to lie in the lowest Landau level, which leads to

$$F = \left[a_0(T - T_{c0}) + \frac{\sqrt{K_4K_1}}{l^2}\right]|\Psi_x|^2 + \left[a_0(T - T_{c0}) + \frac{\sqrt{K_4(K + K_1)}}{l^2}\right]|\Psi_y|^2 - ibM_s(\Psi_x\Psi_y^* - \Psi_y\Psi_x^*)$$

and $l = \sqrt{\hbar c/2eH}$. This quadratic form is easily diagonal-

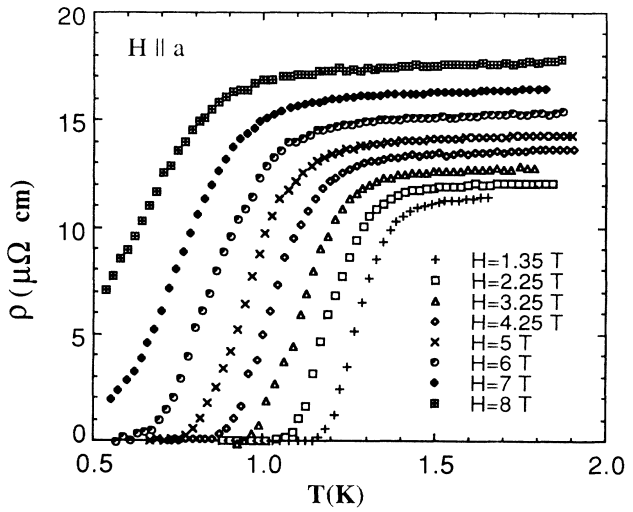

FIG. 2. Resistance vs temperature below 2 K in various magnetic fields along the tetragonal **a** axis of URu_2Si_2.

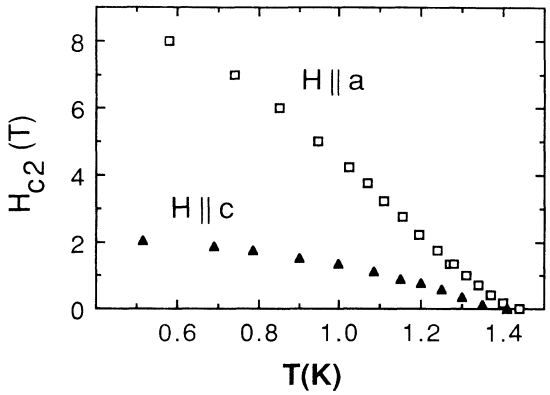

FIG. 3. Upper critical field vs temperature for the field along the tetragonal **a** and **c** axis in URu_2Si_2.

ized. Setting the lowest eigenvalue equal to zero in the usual fashion gives $H_{c2}(T)$. The relation is most easily written in implicit form

$$[a_0(T-T_{c0})+\frac{e}{\hbar c}\sqrt{K_4}(\sqrt{K_1}+\sqrt{K_1+K})H]^2 = \left[\frac{e}{\hbar c}\sqrt{K_4}(\sqrt{K_1}-\sqrt{K_1+K})\right]^2 H^2+b^2M_s^2. \quad (1)$$

At $T=T_c$, one finds a slope

$$\left.\frac{dH_{c2}}{dT}\right|_{T_c} = \frac{-a_0\hbar c/e}{\sqrt{K_4}(\sqrt{K_1+K}+\sqrt{K_1})} = -S_1,$$

whereas when H_{c2} is large enough that bM_s can be neglected, we find

$$\frac{dH_{c2}}{dT} \rightarrow \frac{-a_0\hbar c/e}{\sqrt{K_4}(\sqrt{K_1+K}+\sqrt{K_1}-|\sqrt{K_1+K}-\sqrt{K_1}|)} = -S_2.$$

There is a region of low slope near T_c which crosses over to a higher slope at lower temperatures. Physically, the form of Ψ very near T_c is determined by the coupling to M_s with its accompanying slope dH_{c2}/dT. At higher fields (lower T), the form of Ψ is determined by the direct coupling to H as opposed to M_s, which will tend to give a larger slope. *Hence the curvature is always upward.*

Equation (1) may be rewritten as

$$\left[(T-T_{c0})+\frac{H}{S_1}\right]^2 = \left[\frac{1}{S_1}-\frac{1}{S_2}\right]^2 H^2+(T_c-T_{c0})^2, \quad (2)$$

which we have fitted to our data as shown in Fig. 4. With $T_c=1.44$ K, a three-parameter fit yields

$$S_1 = -dH_{c2}/dT|_{T\rightarrow T_c} = 4.12 \text{ T/K},$$

$$S_2 = -dH_{c2}/dT|_{T\ll T_c} = 11.90 \text{ T/K},$$

and $T_{c0}=1.38$ K. The shift of 0.06 K of T_c from T_{c0} is a measure of the strength of this coupling. Note that the fit is very good near T_c where Ginzburg-Landau theory is expected to be valid.

When H is along $\hat{\mathbf{z}}$ (tetragonal **c** axis), the solution $\Psi_-=0$ is favored by both the external field and the coupling to M_s. Hence there is no competition in determining the form of Ψ and no upward curvature in $H_{c2}(T)$ near T_c, as observed. The H_{c2} curve is simply shifted by the coupling to $\mathbf{M}_s$.

It is difficult to find other explanations for the pattern of upward curvature which we observe in URu_2Si_2. Within standard Ginzburg-Landau and Werthamer-Helfand-Hoenberg[13] theories, there is no mechanism for upward curvature near T_c. Such an effect can occur in low-dimensional systems if there exists a dimensional-crossover effect. However, dimensional crossover cannot apply to URu_2Si_2 where the coherence length is much larger than the unit-cell dimensions. Sample-dependent explanations like inhomogeneities and weak surface superconductivity can be excluded by the existence of upward curvature for only one field direction.

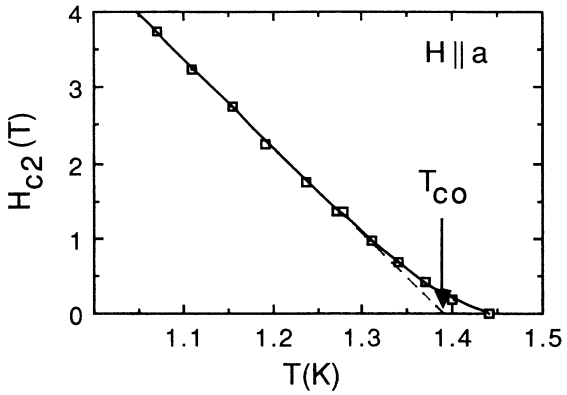

FIG. 4. Fit to the upper critical field below 4 T. The open squares are the experimental data, and the solid line is the fit to Eq. (2). T_{c0} is shown as the extrapolation of the dashed line to zero field.

Some conclusions can now be drawn about the overall phase diagram of URu_2Si_2 in the H-T plane, following the discussion in Ref. 10, and using the notation for the phases defined there. Precisely at T_c and $H=0$, the system must be in the A phase. The apparent absence of two zero-field specific-heat anomalies implies that this phase is then realized along the $H=0$ line down to $T=0$, and most likely is the only superconducting phase for $H\|c$. This is consistent with recent torsional oscillator measurements.[14] Reference 10 then predicts that there must be two phases for H in the basal plane: A at low fields and C at high fields.

In summary, we have shown how upward curvature in the upper critical field of URu_2Si_2 can originate from the coupling of a multicomponent superconducting order parameter with an ordered antiferromagnetic moment. Unusual features have also been observed in the shape of the upper critical field in UPt_3 and U_6Fe (Refs. 8, 15–17). These may also be explained by a theory[18,19] based on a d-wave superconductor suitably modified for the appropriate symmetry. The upward curvature in the upper critical field of URu_2Si_2 and, in particular, the anisotropy of this phenomenon strongly indicates that this low-mass heavy-fermion compound is an unconventional superconductor. This poses the interesting possibility that other materials with effective masses of order 25–30 such as U_6Fe may have multicomponent superconducting order parameters.

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