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Upper critical magnetic fields of pure and thoriated  $UBe_{13}$ 

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We present measurements of the upper critical magnetic field  $H_{c2}(T)$  determined from the dc magnetization of polycrystalline samples of  $U_{(1-x)}Th_xBe_{13}$  with  $x=0$  and  $x=0.0331$  and a single crystal of pure  $UBe_{13}$ . We find changes in slope in the  $H_{c2}$  vs  $T$  phase diagrams of both polycrystalline samples which suggests entrance into a second superconducting phase at about  $T_c/2$ . A degradation of the superconducting critical field is observed in the single crystal which may be related to the absence of second-phase behavior in this sample. Our results are discussed in light of recent experimental and theoretical treatments of second-phase behavior in heavy-fermion superconductors.

It is now generally accepted that the superconducting states of the heavy-fermion superconductors are unusual. The power-law temperature dependence of thermodynamic and transport properties below  $T_c$  suggests that the superconducting energy gap vanishes at points or lines on the Fermi surface.<sup>1</sup> Further evidence is provided by the unusual shapes of the upper critical magnetic field  $H_{c2}(T)$  which have been difficult to explain in terms of conventional theory.<sup>2</sup> The appearance of a second transition  $T_{cb}$  below the superconducting transition  $T_{ca}$  for  $U_{(1-x)}Th_xBe_{13}$  with  $x$  between 0.02 and 0.04 has also attracted a great deal of attention.<sup>1</sup> This lower transition has been observed in specific heat,<sup>3</sup> thermal expansion,<sup>4</sup> ultrasonic attenuation,<sup>5</sup> and lower critical magnetic field  $H_{c1}(T)$  (Ref. 6) measurements. Studies of the pressure dependence of  $T_{ca}$  and  $T_{cb}$  suggest that both phases are superconducting.<sup>7</sup> Recent muon spin-relaxation experiments suggest that the lower transition is accompanied by the onset of antiferromagnetism.<sup>8</sup> A similar situation appears to exist in pure  $UBe_{13}$ .<sup>9</sup> Recent measurements of  $H_{c1}(T)$  and  $H_{c2}(T)$  display features at about  $T_c/2$  which appear to be related to a "bump" in the specific heat.<sup>10</sup> A sizable ultrasonic attenuation peak observed just below  $T_c$  in  $UBe_{13}$  (Ref. 11) supports the identification of the higher-temperature phase in pure  $UBe_{13}$  with the lower-temperature phase of the thoriated systems.

Rauchschwalbe and co-workers have proposed a model for  $U_{(1-x)}Th_xBe_{13}$  in which a second-order parameter appears at the lower transition  $T_{cb}$  and coexists with the order parameter associated with  $T_{ca}$ . The two order parameters are connected with different portions of the Fermi surface. The  $H_{c2}(T)$  curve one measures is that corresponding to the dominant order parameter at a given tem-

perature. Recently, Kumar and Wölfle<sup>12</sup> have shown that a system which allows pairing in two even-parity states of angular momentum (such as  $s$ - and  $d$ -wave states, for example) will have thermodynamic properties such as  $H_{c2}(T)$  which resemble those observed in  $U_{(1-x)}Th_xBe_{13}$ . They identify  $T_{ca}$ , in the thoriated systems, as the onset of  $s$ -wave pairing with a second-order phase transition at  $T_{cb}$  signaling the onset of  $d$ -wave pairing. The  $d$ -wave transition is suppressed by impurities to a temperature below the  $s$ -wave transition. The situation is reversed in pure  $UBe_{13}$  because of the absence of thorium. In this case,  $T_c$  corresponds to the appearance of the  $d$ -wave state, the order parameter of the  $s$ -wave state being switched on continuously, reaching a significant amplitude at  $T_{cb}$ . This picture explains the absence of a second peak in the specific heat.

In this Rapid Communication, we present measurements of  $H_{c2}(T)$  on polycrystalline samples of  $U_{(1-x)}Th_xBe_{13}$  with  $x=0$  and 0.0331 and a single crystal of pure  $UBe_{13}$ .  $H_{c2}$  is determined from the dc magnetization of the samples using an *in situ* capacitive magnetometer which has been described by Brooks *et al.*<sup>13</sup> The magnetometer is positioned about 2 cm above magnetic center in the mixing chamber of a top-loading dilution refrigerator. Our thermometer is a carbon resistor which is mounted about 4 mm below the sample. The magnetoresistance is corrected for using the phenomenological function of Naughton *et al.*<sup>14</sup> Temperature, magnetization, and magnetic field strength are continuously recorded by a computerized data acquisition system.

The magnetic field is swept back and forth across the transition while the mixing chamber is slowly warmed at about 50  $\mu K/s$  by a heater about 40 cm above the sample.

Warming or cooling at this rate reproduces the same  $H_{c2}(T)$  curve. We have also taken data by cooling well below  $T_c$  in zero field, increasing the field to the desired value and then slowly warming at about  $150 \mu\text{K/s}$  through  $T_c$ . Hereafter, we will refer to data taken by the former method as " $M(H)$ " and the latter as " $M(T)$ ." Both  $M(H)$  and  $M(T)$  data display hysteresis in the superconducting state which can be attributed to flux pinning effects.<sup>15</sup>

We define  $H_{c2}$  to be the point at which flux pinning (and therefore, hysteresis) disappears in a transition to the normal state at fixed  $T$  and increasing  $H$  or vice versa. These transitions are typically less than 0.2 T and 20 mK for  $M(H)$  and  $M(T)$  data, respectively. Simultaneous measurements of resistance and magnetization using this technique<sup>13</sup> indicate that these transitions occur at the onset of finite resistance. We therefore expect our values of  $H_{c2}$  to be somewhat lower than those determined by the midpoint of resistive transitions. The sharpness of the transitions avoids any difficulty associated with large (about 3 T in this case) resistive transition widths.

Our results on the polycrystalline sample of  $\text{UBe}_{13}$  are displayed in Fig. 1. The points represent  $M(H)$  data taken with both superconducting and resistive magnets. The solid lines are guides to the eye. The low-temperature  $H_{c2}(T)$  is linear and extrapolates to  $H_{c2}(0) = 13.0$  T, as the temperature is increased a region of positive curvature is observed. This behavior was seen previously by Rauchschalbe *et al.* who interpret the inflection in  $H_{c2}(T)$  as the emergence of a low-temperature " $a$  phase" out of the high-temperature " $b$  phase".<sup>9</sup> The low-temperature phase corresponds to the high-temperature phase in the thoriated sample as discussed above. In keeping with this view the dashed lines in Fig. 1 are smooth extrapolations of the solid line towards  $H_{c2}(T) = 0$  and  $H_{c2}(0)$  for the  $a$  and  $b$  phases respectively. Our results are consistent, though lower as expected, with earlier resistive measure-

ments on the same sample<sup>16</sup> which are somewhat sparse in the region of the transition.

Our data on the single crystal of  $\text{UBe}_{13}$  are shown in Fig. 2. The crystal is oriented with the field approximately normal to one of the cubic faces. The solid circles represent  $M(H)$  data taken with a resistive magnet. The solid line through the data represents a fit to theory discussed below and extrapolates to  $H_{c2}(0) = 7.8$  T. The qualitative behavior of  $H_{c2}(T)$  for this sample is quite different from that of the polycrystal: neither an inflection point nor any sign of positive curvature is observed. This single crystal was grown in an aluminum flux and aluminum inclusions may degrade the intrinsic  $\text{UBe}_{13}$  behavior.<sup>17</sup> Hence one might expect that these inclusions could also suppress the  $a$ -phase/ $b$ -phase behavior. Should this be the case the effect of aluminum impurities is clearly different from that of thorium. Preliminary attempts to measure an anisotropy in  $H_{c2}(T)$  by varying the orientation of the crystal with respect to the external field indicate that the anisotropy, if it exists, is less than few tenths of a tesla. This is clearly insufficient to explain the 4 T difference in  $H_{c2}(0)$  between the single crystal and the polycrystal. The dashed line above the data represents  $H_{c2}(T)$  for this sample as determined from the midpoints of resistive transitions.<sup>18</sup>

We attempted to fit our single-crystal data to the standard theory of type-II superconductors in the dirty limit.<sup>19</sup> Fixing  $dH_{c2}/dT|_{T=T_c} = -42$  T/K as determined by Maple *et al.*<sup>18</sup> and setting the  $g$  factor  $g_0 = 2$  we found that varying  $T_c$  and the spin-orbit scattering parameter  $\lambda_{s.o.}$  to fit the high-temperature data caused the low-temperature data to exceed our fit by about 0.5 T. The failure of the standard theory is not surprising since it assumes that the normal-state parameters are independent of  $T$  and  $H$  which is not justified in the  $\text{U}_{(1-x)}\text{Th}_x\text{Be}_{13}$  system.<sup>1</sup> An extension of the standard theory to include a phenomenological field dependence of the normal-state parameters

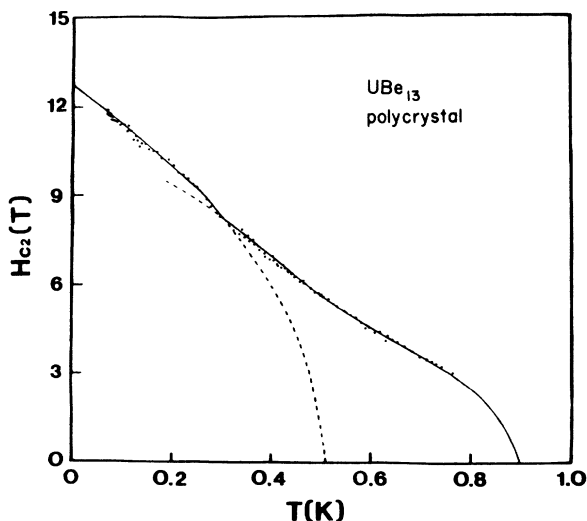


FIG. 1. The upper critical magnetic field of polycrystalline  $\text{UBe}_{13}$ . The solid lines are guides to the eye. The dashed lines are smooth extrapolations of the solid lines as discussed in the text.

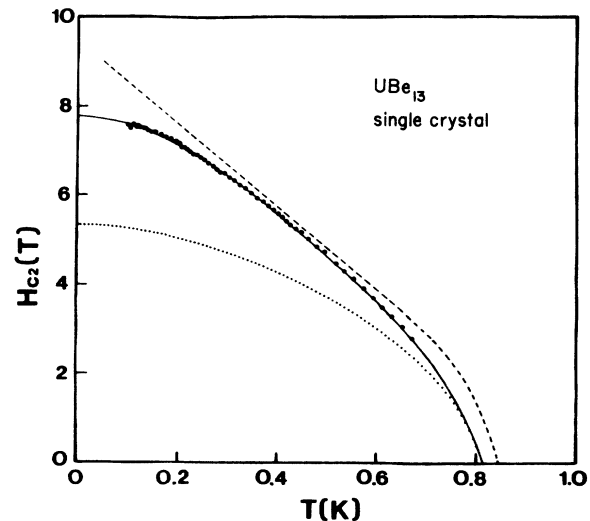


FIG. 2. The upper critical magnetic field of single crystal of  $\text{UBe}_{13}$ . The solid line is a fit to theory from which the dotted line is a particular limit (see text). The dashed line represents earlier resistive measurements of  $H_{c2}(T)$  (Ref. 18).

has been discussed by Tachiki, Koyama, and Takahashi.<sup>20</sup> In their theory, the normal phase is described as a coherent Kondo state in which the condensation energy is comparable to the energy of the magnetic field at  $H_{c2}(T)$ . The field dependence can be introduced by replacing  $T_c \rightarrow T_{c0} \exp(AH^2)$ ,  $\lambda_{s.o.} \rightarrow \lambda_{s.o.}/(1-BH^2)$ , and  $g_0 \rightarrow g_0(1+CH^2)$  in the standard model (see Eq. 3.46 of Ref. 19) where  $A$ ,  $B$ , and  $C$  are constants. Since the magnetic susceptibility of normal  $U\text{Be}_{13}$  has been observed to be nearly independent of magnetic field at low temperatures,<sup>21</sup> we set  $C=0$ ,  $g_0=2$ , and  $dH_{c2}/dT|_{T=T_c} = -42$  T/K as above and fit our data treating  $T_{c0}$ ,  $\lambda_{s.o.}$ ,  $A$ , and  $B$  as adjustable parameters. The result is the solid line in Fig. 2 given by  $T_{c0}=0.813$  K,  $\lambda_{s.o.}=8.13$ ,  $A=6.51 \times 10^{-3}$  ( $\text{T}^{-2}$ ) and  $B=9.30 \times 10^{-3}$  ( $\text{T}^{-2}$ ). The dotted line in Fig. 2 shows the same fit with  $A=B=C=0$ . These results are physically reasonable within the context of the phenomenological theory but the field dependence implied must be verified by experiment or a calculation from first principles.

Our results for the polycrystalline sample of  $U_{(1-x)}\text{Th}_x\text{Be}_{13}$  with  $x=0.0331$  are shown in Fig. 3. The small solid circles and large open circles represent  $M(H)$  and  $M(T)$  data, respectively, both taken using a superconducting magnet. The solid line represents a theoretical fit discussed below. A discontinuity in slope of the  $M(H)$  data at 0.25 K and 3.9 T marks the transition from the high-temperature  $a$  phase to a lower temperature  $b$  phase. An expanded view of this region is shown in the inset of Fig. 3 which displays the difference between all the  $M(H)$  data and a linear least-squares fit to the  $M(H)$   $a$ -phase data above 0.25 K. The transition temperature and initial slope of the  $a$  phase are in good agreement with earlier resistive measurements.<sup>16</sup> This agreement justifies our fixing  $dH_{c2}/dT|_{T=T_c}$  of the pure single crystal to the value determined by Maple *et al.*<sup>18</sup> Preliminary measurements indicate that the low-temperature linearity of  $H_{c2}(T)$  in the  $b$  phase persists to at least 80 mK, the results shown extrapolate to  $H_{c2}(0) = 5.4$  T.

The solid line in Fig. 3 is the result of a fit of the  $a$ -phase data to the standard theory of type-II superconductors in the dirty limit.<sup>19</sup> We set  $g_0=2$ ,  $dH_{c2}/dT|_{T=T_c} = 32$  T/K (Ref. 16), and treated  $\lambda_{s.o.}$  and  $T_c$  as adjustable parameters. The results are  $T_c=0.642$  and  $\lambda_{s.o.}=10.28$ . Although the accessible portion of  $H_{c2}(T)$  for the  $a$  phase can be described by the standard theory it is not clear that this is an appropriate choice. Assuming the  $a$  phase of the thoriated sample to be similar to that of the pure polycrystal we would expect  $H_{c2}(T)$  to be linear at low temperatures.

In conclusion, we have determined  $H_{c2}(T)$  from the dc magnetization of polycrystalline samples of  $U_{(1-x)}\text{Th}_x\text{Be}_{13}$  with  $x=0$  and 0.0331, and single-crystal sample with  $x=0$ . Both polycrystalline samples display features in  $H_{c2}(T)$  which can be interpreted as phase transitions to another superconducting state at temperatures of about

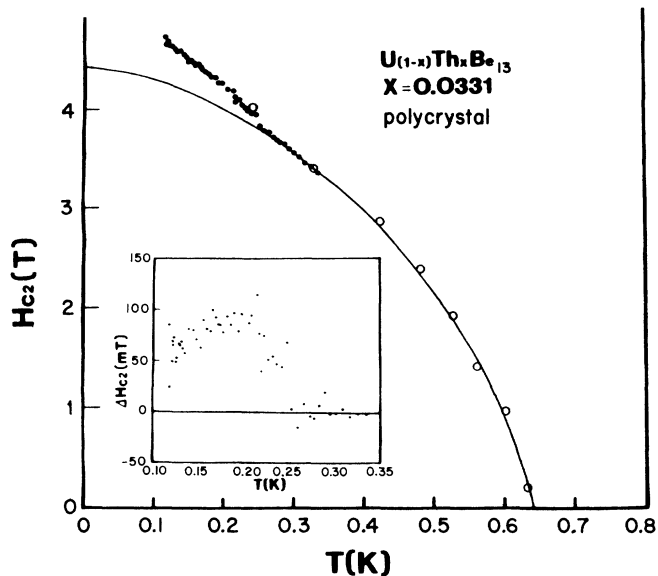


FIG. 3. The upper critical magnetic field of a polycrystalline sample of  $U_{(1-x)}\text{Th}_x\text{Be}_{13}$  with  $x=0.0331$ . The solid line is a fit of the high-temperature data to theory (see text). The inset shows an expanded view of the low-temperature data with a linear fit subtracted (see text).

$T_c/2$ . The temperature dependence of  $H_{c2}$  observed for the polycrystal with  $x=0$  is linear at low temperatures and displays positive curvature at higher temperatures in agreement with the earlier work of Rauchschalbe *et al.*<sup>2</sup> The temperature dependence of  $H_{c2}$  for the single-crystal sample can be described by a phenomenological extension of the standard theory of type-II superconductors in the dirty limit. Our low-temperature results for the single crystal, indicating a saturation of  $H_{c2}(T)$  as  $T \rightarrow 0$ , are qualitatively different from earlier resistive measurements on the same sample. The temperature dependence of the  $a$  phase of the thoriated sample can be described by the standard theory.

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