Upper critical magnetic field of the heavy-electron superconductors $U_{1-x}Th_xBe_{13}$ ($x=0$ and 2.9%) doped with paramagnetic Gd and other rare-earth ions

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(Received 1 May 1990)

The temperature T dependence of the upper critical magnetic field H_{c2} of the heavy-electron superconductors $U_{1-x}Th_xBe_{13}$ ($x=0$ and 2.9%) doped with various concentrations of Gd has been determined from low-frequency ac magnetic susceptibility measurements in magnetic fields up to 60 kOe. The $H_{c2}(T)$ curves for $U_{1-x}Gd_xBe_{13}$ samples deviate from the $H_{c2}(T)$ curves of UBe_{13} near T_c, which is consistant with the depairing of superconducting electrons via the Zeeman interaction between the spins of the superconducting electrons and the exchange field associated with the Gd spins. This suggests that UBe_{13} exhibits singlet superconductivity. In contrast, the $H_{c2}(T)$ curves for $(U_{0.97}Th_{0.03})Be_{13}$ doped with Gd scale with $H_{c2}(T)$ of pure $(U_{0.97}Th_{0.03})Be_{13}$ and do not reflect superconducting depairing by the Gd ions. These results are consistent with either strong spin-orbit scattering due to the presence of Th in UBe_{13}, or to a qualitatively different type of superconductivity involving triplet spin pairing in $(U_{0.97}Th_{0.03})Be_{13}$. Measurements of the temperature dependence of H_{c2} for $U_{1-x}R_xBe_{13}$ compounds where $R=$La, Lu, and Ce for various compositions x as well as $U_{0.985}R_{0.015}Be_{13}$ compounds for $R=$Tb, Dy, Ho, and Er are also presented and compared with the Gd-doped UBe_{13} system. The results of low-temperature specific-heat measurements of UBe_{13} doped with various concentrations of Gd are also discussed.

INTRODUCTION

The upper critical magnetic field $H_{c2}(T)$ of the heavy-electron superconductor UBe_{13} ($T_c \lesssim 1$ K) is characterized by several peculiarities. First, the magnitude of the initial slope is very large ($\sim$420 kOe/K) and is the highest value that has been reported to date for a bulk superconductor. Second, $H_{c2}(T)$ is practically linear below $T\approx 0.7$ K and down to 50 mK. Third, $H_{c2}(0)$ exceeds the paramagnetic limit by an order of magnitude.[1] Measurements on polycrystalline samples, however, show an upturn in $H_{c2}(T)$ below $T_c/2$.[2] The anomalous superconducting properties of UBe_{13} and other heavy-electron superconductors have led to the speculation that these materials may exhibit an unconventional type of superconductivity similar to the triplet superfluidity displayed by liquid ^{3}He below a few mK.

These materials, which are characterized by enormous normal-state electronic specific-heat coefficients γ, as large as $\sim$1 J/mol K^2, have relatively low superconducting transition temperatures $T_c \lesssim 1$ K and coherence lengths $\xi \approx 50$ Å, extraordinarily large upper critical magnetic fields H_{c2} with unusual temperature dependences, and power-law dependences in T for $T \ll T_c$ of the specific heat,[3] ultrasonic attenuation coefficient,[4] thermal conductivity,[3] NMR spin-lattice relaxation rate,[5] and magnetic-field penetration depth.[6] The absence of an exponential activation energy, predicted by the BCS theory for a conventional superconductor, in these quantities indicates that the energy gap is anisotropic and vanishes at points or lines on the Fermi surface.[7] In analogy with the superfluid A phase of ^{3}He, an anisotropic order parameter in these materials suggests an interparticle interaction in partial waves with angular momentum $l \neq 0$.[8] Since a fermion wave function must be antisymmetric under exchange of any two particles, a pair of total spin $S=0(1)$ must condense into an even- (odd-) l state. The most compelling evidence for unconventional superconductivity in heavy-electron materials is provided by the presence of two distinct superconducting transitions in UPt_3 and (U,Th)Be_{13}.[9,10] This splitting of the superconducting transition is consistent with a non-s-wave pairing.

Because of strong spin-orbit coupling, the rotational invariance in spin space is missing in heavy-electron compounds, and therefore one should refer to the spatial inversion parity of the order parameter: even (for $S=0$, singlet) and odd (for $S=1$, triplet). For convenience, we will in some cases use the terminology of triplet and singlet pairing for odd or even parity of the order parameter. A complete classification of the possible odd- and even-parity superconducting states for the cubic crystalline symmetry of UBe_{13} based on group-theoretical arguments was worked out by Volovik and Gor'kov.[11] They found that in odd-parity states the energy gap vanishes only at points on the Fermi surface, whereas in even-parity states lines of zeros are also possible. As a consequence, if the heat capacity C below T_c has a T^2 dependence, triplet pairing is excluded, whereas a T^3 dependence is possible for both triplet and singlet pairing. The symmetry classification of the superconducting-state order parameter can be determined, without any indication of its parity, however, by measuring the anisotropy of H_{c2} near T_c.[12] It was suggested that the pairing mechanism in heavy-electron compounds could not be due to phonons because in these materials the Coulomb repulsion is more effective than the phonon-mediated electron-electron interaction.[13] A number of authors[14] have proposed the exchange of antiferromagnetic spin fluctuations as the mechanism leading to Cooper pairing in heavy-electron compounds.

Magnetic as well as normal impurity ions have proven to be very effective in suppressing superconductivity in UBe_{13}.[15] Particularly interesting behavior is observed when nonmagnetic Th is substituted for U to form the $U_{1-x}Th_xBe_{13}$ system. With increasing x, T_c exhibits, initially, a nearly linear decrease with a distinct minimum at $x \approx 1.72\%$, then a broad maximum at $x \approx 2.5\%$, with a subsequent decrease at higher concentrations. For compositions x between $\sim 2\%$ and $\sim 4\%$, two transitions have been observed in the specific heat[10] below 1 K, the first one, at T_{c1}, associated with the development of the superconducting state and the second one, at a lower temperature T_{c2}, corresponding to another phase transition that occurs without destroying superconductivity. Two possible explanations for the lower peak are that (a) it is associated with a second superconducting phase with a different order-parameter symmetry in analogy with the two phases of superfluid ^{3}He, and (b) that it is due to the formation of an antiferromagnetic state which coexists with superconductivity. Batlogg *et al.*[16] observed strong ultrasonic attenuation with a peak near the second transition which they interpreted as evidence for antiferromagnetic ordering. Recent muon-spin relaxation measurements in $U_{0.965}Th_{0.035}Be_{13}$ by Heffner *et al.*[17] confirmed the onset of weak magnetism (10^{-3}–$10^{-2}\mu_B$/U atom) at T_{c2}. Measurements of the temperature dependence of the lower critical field $H_{c1}(T)$ in the $U_{1-x}Th_xBe_{13}$ series yielded some rather complicated results: For $x = 0\%$, H_{c1} has a quadratic temperature dependence; for $x = 1.0\%$, $|dH_{c1}/d(T^2)|$ decreases abruptly below T_{c2} even though specific-heat data do not show any second transition; and in the region where the specific-heat data show two anomalies ($x \geq 2\%$), $|dH_{c1}/d(T^2)|$ increases below T_{c2}.[18,19] Rauchschwalbe *et al.*[18] suggested that the increase in slope is related to a second superconducting transition at T_{c2} with a different order-parameter symmetry. However, magnetic order can also lead to such behavior in $H_{c1}(T)$.[17] From measurements of T_c under pressure, we found evidence for two superconducting states in the $U_{1-x}Th_xBe_{13}$ system, one type of which occurs at $x \lesssim 1.72\%$ and another at $x \gtrsim 1.72\%$, at atmospheric pressure.[20] Consistent with these findings are the results of muon-Knight-shift measurements[21] on $U_{1-x}Th_xBe_{13}$ samples with $x = 0$ and 3.3 %, which show a nearly conventional superconducting Knight shift for $x = 0\%$ and no deviation from the normal-state Knight shift for $x = 3.3\%$ below T_c.

In a first part of this paper, we report the results of measurements of $H_{c2}(T)$ for $U_{1-x}Th_xBe_{13}$ compounds with $x = 0$ and 2.9 % doped with various concentrations of Gd. The objective of this experiment was to see how the $H_{c2}(T)$ curve of $U_{1-x}Th_xBe_{13}$ ($x = 0$ and 2.9 %) is modified by the introduction of Gd^{3+} ions, in order to obtain information about the relative orientation of the spins within a superconducting electron pair in these two systems. Preliminary accounts of the $H_{c2}(T)$ measurements and their interpretation were reported in Refs. 22 and 23. In the second part, we present the results of measurements of $H_{c2}(T)$ for $U_{1-x}R_xBe_{13}$, where R represents magnetic as well as nonmagnetic rare-earth ions for various values of x with particular interest in the behavior of $H_{c2}(T)$ in low magnetic fields. Low-temperature specific-heat data are also presented, in the third and final part, for pure UBe_{13} and UBe_{13} doped with various concentrations of Gd.

EXPERIMENTAL DETAILS

The arc-melted polycrystalline samples used in this investigation were prepared in a manner described previously.[15] The upper critical magnetic field $H_{c2}(T)$ was determined from the ac magnetic susceptibility χ_{ac}, measured on bulk specimens by means of a standard mutual inductance technique at a frequency of 16 Hz in a ^{3}He-^{4}He dilution refrigerator. The temperature was varied between 70 mK and 1 K in fixed magnetic fields up to 60 kOe that were produced by a superconducting solenoid. The specific-heat data were taken in a ^{3}He semiadiabatic calorimeter using the heat-pulse technique. Sharp superconducting transitions were generally observed in $H = 0$ Oe, broadening somewhat in applied magnetic fields. However, for some compositions x in the La-, Lu-, and Ce-doped materials, the situation is complicated by the presence of small secondary superconducting transitions at higher temperatures. In these cases a reasonable estimate is made of the total χ_{ac} change due to the larger transition and the higher-temperature transition is ignored. The value of T_c is defined as the temperature at which the superconducting change $\Delta\chi_{ac}$ in the ac magnetic susceptibility is 10% of its full value in $H = 0$ Oe.

RESULTS

Typical $\chi_{ac}(T)$ curves for a $U_{1-x}Gd_xBe_{13}$ sample with $x = 0.228\%$ and a $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$ sample with $x = 0.500\%$ are shown in Figs. 1 and 2, respectively. Displayed in Fig. 3 are the $H_{c2}(T)$ curves for $U_{1-x}Gd_xBe_{13}$ ($x = 0$, 0.228, 0.543, 1.018, 1.506, and 1.987%) and $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$ ($x = 0$, 0.291, 0.500, 0.772, and 1.050%). The data for UBe_{13} are taken from a previous publication.[1] The initial slope of the upper critical field H_{c2} in this work was determined from the data above 2 kOe in $U_{1-x}R_xBe_{13}$ and from the data above 1 kOe in $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$.

The $H_{c2}(T)$ curves of Gd-doped UBe_{13} samples deviate strongly from that of the pure compound, especially for $x > 1.018\%$, where they show bending in high fields and a rapid decrease of $H_{c2}(0)$, similar to what is seen in con-

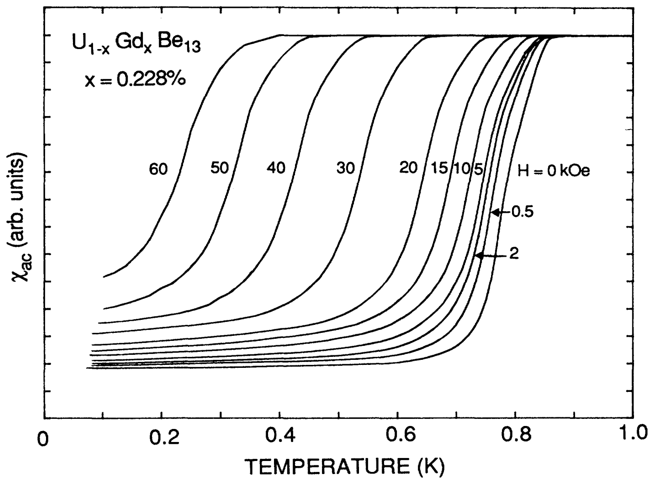

FIG. 1. ac magnetic susceptibility χ_{ac} vs temperatures in various applied magnetic fields for a $U_{1-x}Gd_xBe_{13}$ compound with $x=0.228\%$. For clarity, some low-magnetic field transition curves are not shown.

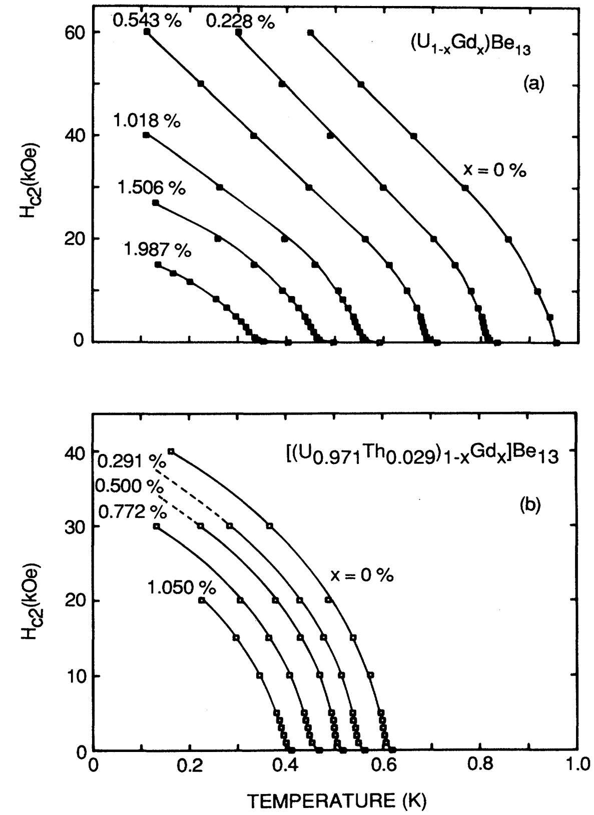

FIG. 3. Upper critical magnetic field H_{c2} vs temperature determined from $\chi_{ac}(T,H)$ measurements for (a) $U_{1-x}Gd_xBe_{13}$ compounds with $x=0$, 0.228, 0.543, 1.018, 1.506, and 1.987 %, and (b) $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$ with $x=0$, 0.291, 0.500, 0.772, and 1.050 %. Solid lines are guides to the eye.

ventional superconductors doped with magnetic ions. For $x \leq 1.018\%$, the overall shape and, in particular, the linear T dependence of $H_{c2}(T)$ above ~ 20 kOe resembles that of the pure UBe_{13} compound. A peculiar "foot," whose size increases with x, develops in the low-magnetic-field region ($H < 2$ kOe). The initial slope decreases at a rate of 160 kOe/at. % and the critical field $H_{c2}(0)$ by 50% when $x=1.018\%$, as summarized in Table I. The "foot" near T_c in the $H_{c2}(T)$ curves for $U_{1-x}Gd_xBe_{13}$ is apparently due to the exchange field H_J associated with the Gd^{3+} ions. In fact, as the temperature is lowered, H_J, which is proportional to the Gd spin magnetization M, grows quickly and the paramagnetic reduction of H_{c2} is strong in low fields. This reduction of H_{c2} continues until M and, in turn, H_J and its pair-breaking effects saturate, at which point the behavior characteristic of UBe_{13} is recovered. It is noteworthy that the critical field $H_{c2}(0)$, which is larger than the paramagnetic limiting field H_p in UBe_{13}, becomes smaller than H_p for $U_{1-x}Gd_xBe_{13}$ compounds with Gd concentrations $x > 1.1018\%$.

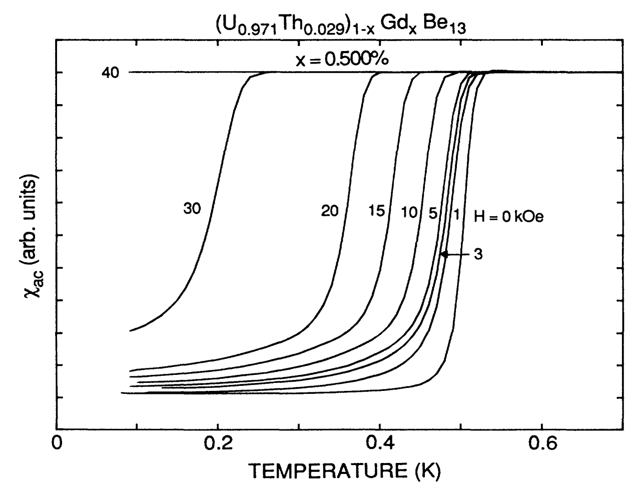

FIG. 2. ac magnetic susceptibility χ_{ac} vs temperature in various applied magnetic fields for a $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$ compound with $x=0.500\%$. For clarity, some low-magnetic-field transition curves are not shown.

In contrast, the shape of the $H_{c2}(T)$ curves of Gd-doped $(U_{0.971}Th_{0.029})Be_{13}$ remains unchanged for concentrations $0\% \leq x \leq 1.050\%$, indicating that the superconducting electron pairs are rather indifferent to the exchange field H_J associated with the Gd spins. All curves can be well described just by scaling the one for $x=0$ with the corresponding T_c. The initial slope decreases with increasing x, and the critical field $H_{c2}(0)$ drops by 30% when x increases from 0% to 1.050%, as summarized in Table II.

In order to shed more light on the low-field behavior of the $U_{1-x}Gd_xBe_{13}$ system, we have also measured the upper magnetic critical field in $U_{1-x}R_xBe_{13}$ compounds where U is partially replaced by other magnetic as well as nonmagnetic rare-earth ions R. Shown in Fig. 4 are the $H_{c2}(T)$ curves for $U_{1-x}Ce_xBe_{13}$ samples with $x=0$, 1.12,

TABLE I. Parameters used to calculate $H_{c2}(T)$, for various concentrations x, of $U_{1-x}Gd_xBe_{13}$ (see text for details) and physical quantities characterizing the superconducting state, estimated from $\chi_{ac}(T,H)$ measurements. For all concentrations, $\mathcal{J}=0.036$ eV, $g_J=2$, $J=\frac{7}{2}$, $\lambda_{so}=10$, and $T_{c0}=0.955$ K.

x (%)	T_c (K)	$H^*_{c2}(0)$ (kOe)	H_J[a] (kOe)	$H_{c2}(0)$ (kOe)	H_p[b] (kOe)	$(-dH_{c2}/dT)_{T_c}$ (kOe/K)
0	0.955[c]	230	0	100	72.2	420[c]
0.228	0.834	204	1.8	87	63.1	400
0.543	0.710	174	4.2	70	53.7	350
1.018	0.592	145	7.9	45	44.8	190
1.506	0.497	122	11.7	32	37.6	160
1.987	0.404	99	15.4	19	30.6	103
2.747	<0.08		21.3			

[a]At saturation.
[b]Calculated from $H_p=1.3\sqrt{\lambda_{so}}H_{p0}$, where H_{p0} (kOe)$=18.4T_c$ (K).
[c]From Ref. 1.

2.24, 2.94, and 3.24%. The effect of Ce on the $H_{c2}(T)$ curves is similar to that of Gd ions in that both $H_{c2}(0)$ and the magnitude of the initial slope decrease quickly with increasing x and attain values of 55 kOe and 380 kOe/K, respectively, at 1.12%. The "foot" observed in low fields in the Gd-doped UBe_{13} is also seen with Ce, but its size is substantially smaller. Shown in Fig. 5 are the $H_{c2}(T)$ data for $U_{0.985}R_{0.015}Be_{13}$ compounds where $R=$Tb, Dy, Ho, and Er. At low temperatures, the curves exhibit bending similar to that induced by Gd and Ce impurities with no particular signature for any rare earth. As noted previously for Gd and Ce impurities, $H_{c2}(0)$ and the initial slope are also depressed. These parameters are summarized in Table III. The positive curvature near T_c is also present in the $H_{c2}(T)$ curves of $U_{0.985}R_{0.015}Be_{13}$ compounds. When nonmagnetic La and Lu trivalent ions are substituted for U, the effect on $H_{c2}(T)$ is perhaps even stronger than their magnetic counterparts. Representative data for $U_{1-x}Lu_xBe_{13}$ ($x=0$, 0.6, 1, and 1.6%) are shown in Fig. 6. The linear behavior of $H_{c2}(T)$ for $x=0\%$ is modified, and the critical field $H_{c2}(0)$ is reduced from 100 kOe in UBe_{13} to 15 kOe in $U_{0.964}La_{0.036}Be_{13}$ and 10 kOe in $U_{0.984}Lu_{0.016}Be_{13}$. It is noteworthy that the effect of Lu impurities on the heavy-electron state in UBe_{13} is much stronger than that of La impurities as can be seen from the value of the initial slope of H_{c2}. In conventional superconductors, small quantities of nonmagnetic impurities are not expected to change significantly the shape of the upper critical field. The "foot-shaped" feature observed in low fields for compounds containing magnetic rare-earth impurity ions is also present for La and Lu impurities, but its size is smaller.

The concept of exchange field does not apply in $U_{1-x}La_xBe_{13}$ and $U_{1-x}Lu_xBe_{13}$ since trivalent La and Lu ions are nonmagnetic and thus $H_J=0$. Consequently, another mechanism must be responsible for the small feature near T_c and the high-field shape of H_{c2}, which is reminiscent of pair breaking. It has been suggested[24] that the replacement of a Kondo scattering center (i.e., U) by a nonmagnetic impurity, such as La and Lu, induces a magnetic moment at that site in the coherent state, essentially because there is a conduction-electron compensation cloud without a local spin moment. Therefore, the situation would be equivalent to that of magnetic impurities.

Estimates of $\Delta T_c/T_c$, where ΔT_c is defined as T_c-T_{c1}, are listed in Table III. The temperature T_{c1} is the T in-

TABLE II. Parameters used to calculate $H_{c2}(T)$, for various concentrations x, of $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$ (see text for details) and physical quantities characterizing the superconducting state, estimated from $\chi_{ac}(T,H)$ measurements. For all concentrations, $\mathcal{J}=0.036$ eV, $g_J=2$, $J=\frac{7}{2}$, $\lambda_{so}=10$, and $T_{c0}=0.609$ K.

x (%)	T_c (K)	$H^*_{c2}(0)$ (kOe)	H_J[a] (kOe)	$H_{c2}(0)$ (kOe)	H_p[b] (kOe)	$(-dH_{c2}/dT)_{T_c}$ (kOe/K)
0	0.609	161	0	44	46.1	380
0.291	0.551	146	2.3	41	41.7	360
0.500	0.508	134	3.9	38	38.4	320
0.772	0.457	121	6.0	33.5	34.6	250
1.050	0.406	107	8.2	29	30.7	200

[a]At saturation.
[b]Calculated from $H_p=1.3\sqrt{\lambda_{so}}H_{p0}$.

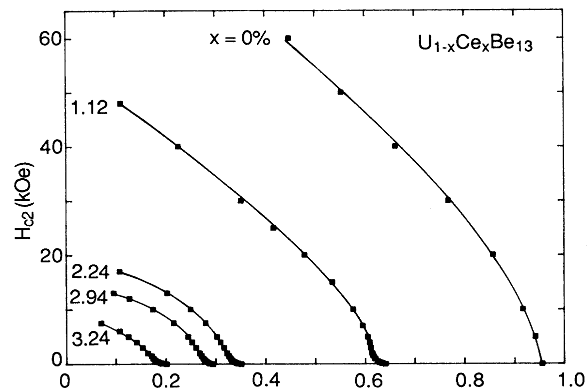

FIG. 4. Upper critical magnetic field H_{c2} vs T temperature determined from $\chi_{ac}(T,H)$ measurements for $U_{1-x}Ce_xBe_{13}$ compounds with $x=0$, 1.12, 2.24, 2.94, and 3.24 %. Solid lines are guides to the eye.

TABLE III. Various superconducting parameters of $U_{0.985}R_{0.015}Be_{13}$ compounds for selected rare-earth R elements estimated from $\chi_{ac}(T,H)$ and $H_{c2}(T)$ data.

R	T_c (K)	$\Delta T_c/T_c$	$H_{c2}(0)$ (kOe)	$(-dH_{c2}/dT)_{T_c}$ (kOe/K)
La	0.570[a]	0.043[a]	38[b]	270[b]
Ce	0.540[a]	0.036[a]	55[c]	380[c]
Nd	0.441			
Sm	0.498[d]			
Gd	0.497	0.062	32	160
Tb	0.440	0.073	24	118
Dy	0.439	0.075	26	140
Ho	0.348	0.082	18	153
Er	0.363	0.064	21	175
Lu	0.230[a]	0.034[a]	10[d]	160[d]

[a]Deduced by interpolation of the existing data.
[b]For $x=1.7\%$.
[c]For $x=1.12\%$.
[d]For $x=1.6\%$.

tercept of the $H_{c2}(T)$ data linearly extrapolated from above 2 kOe as shown in Fig. 7 for $U_{0.985}Ho_{0.015}Be_{13}$. Clearly, $\Delta T_c/T_c$ is larger when R is a rare-earth ion with a partially filled $4f$ electron shell (except Ce) by as much as 50% over that when R is a rare earth with empty or filled $4f$ electron shell. This is consistent with the interpretation of the low-field anomaly in H_{c2} of $U_{1-x}Gd_xBe_{13}$ as resulting from the exchange field associated with the magnetic ions. The data seem to correlate with the effective moment of the rare-earth ions in their Hund's rules ground state, but this may be fortuitous because effects such as crystalline electric-field splitting of the R-ion energy levels or variation of the conduction-$4f$ electron exchange-interaction parameter $\mathcal{J}$ with R could be important in determining the magnitude of the anomaly observed at T_c. The effect of Ce is small and comparable to that of La and Lu.

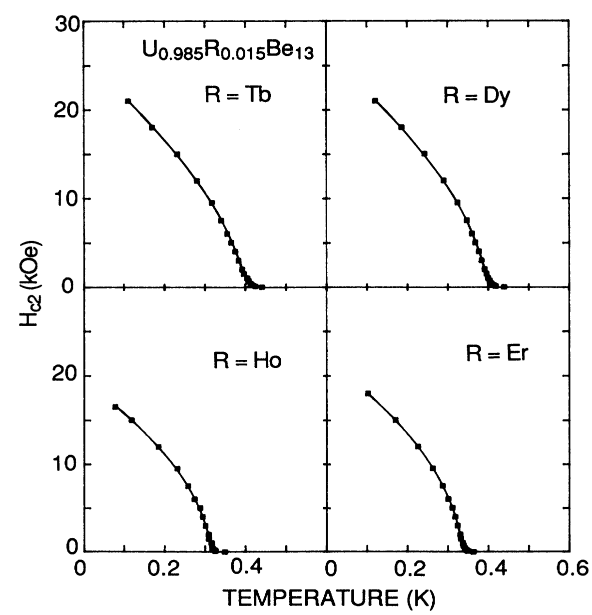

FIG. 5. Upper critical magnetic field H_{c2} vs temperature T determined from $\chi_{ac}(T,H)$ measurements for $U_{0.985}R_{0.015}Be_{13}$ where $R=$Tb, Dy, Ho, and Er. Solid lines are guides to the eye.

Shown in Fig. 8 are the results of specific-heat $C(T)$ measurements taken on $U_{1-x}Gd_xBe_{13}$ samples with $x=0$, 0.228, 1.018, and 1.987 % in the temperature range 0.4 K $< T <$ 22 K. The $C(T)$ data for UBe_{13} show three anomalies: a broad shoulder near 10 K, a Schottky-like anomaly around 2 K, and a jump related to the superconducting transition below 1 K. Incorporation of Gd increases the phonon contribution in UBe_{13}, first rapidly, and then more slowly for higher values of x. The data can be described in the temperature range 12–21 K by the sum of an electronic term ($C_e=\gamma T$) and a lattice term ($C_l=\beta T^3$), i.e.,

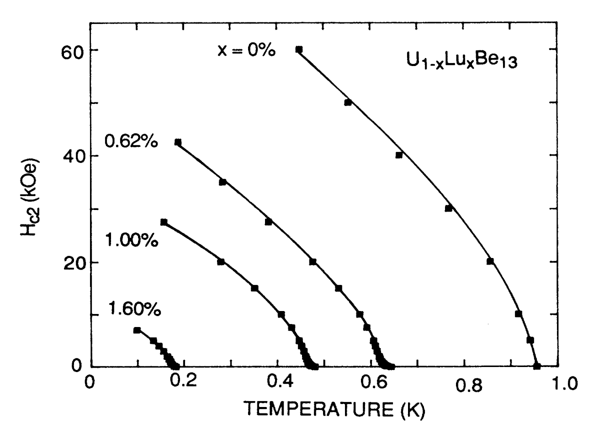

FIG. 6. Upper critical magnetic field H_{c2} vs temperatures T determined from $\chi_{ac}(T,H)$ measurements for $U_{1-x}Lu_xBe_{13}$ compounds with $x=0$, 0.62, 1.00, and 1.60 %. Solid lines are guides to the eye.

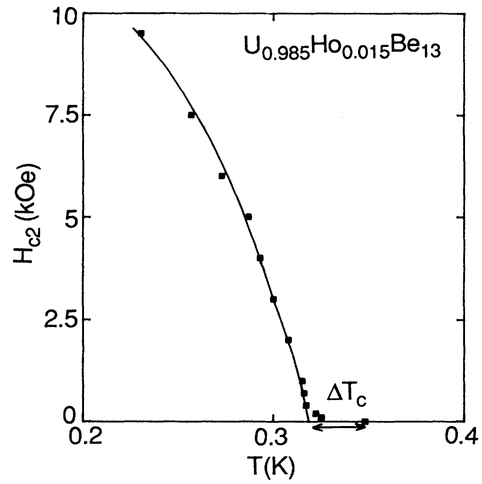

FIG. 7. Low-field H_{c2}-vs-T data for $U_{0.985}Ho_{0.015}Be_{13}$ illustrating how ΔT_c is estimated.

$$C(T)=\gamma T+\beta T^3 \ . \tag{1}$$

The values of γ, β, and θ_D are included in Table IV. The broad shoulder in UBe_{13} near 10 K is unaffected by the Gd impurities, and the Schottky-like anomaly at $T\approx 2$ K does not shift significantly in temperature. A plot of C/T versus T^2 in Fig. 9 shows that γ, the electronic specific-heat coefficient, extrapolated from below 3 to 0 K, decreases with increasing x, initially at a rate of 120 mJ/(mol K^2 at. % Gd), then less rapidly at a rate ≈ 50 mJ/mol K^2 at. % Gd for $x>0.228\%$. The broadened superconducting transitions for $x=0\%$ and 0.228% can be replaced by sharp discontinuities on a C/T-versus-T plot (not shown), such that the entropy involved in the superconducting transition is conserved. The transition temperatures T_c and the specific-heat jump ΔC values determined in this manner are included in Table IV.

DISCUSSION

The multiple pair-breaking theory for a conventional type-II superconductor in the dirty limit[25,26] ($l \ll \xi$) and strong spin-orbit scattering ($\lambda_{so} \gg 1$) was used to de-

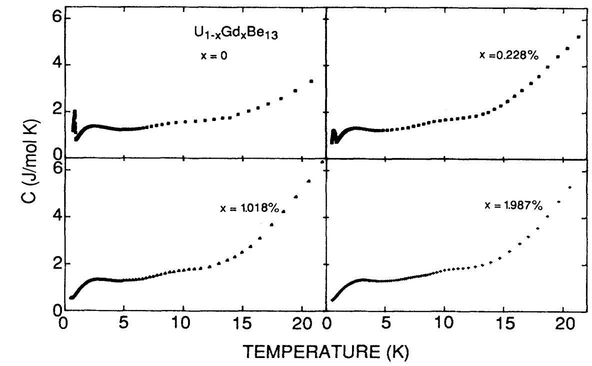

FIG. 8. Specific heat C vs temperature T data for $U_{1-x}Gd_xBe_{13}$ compounds with $x=0$, 0.228, 1.018, and 1.987 % between 0.5 and 22 K.

scribe the H_{c2} curves for the $U_{1-x}Gd_xBe_{13}$ compounds. The expression for $H_{c2}(T)$ in kOe is

$$\begin{aligned} H_{c2}(T) &= H^*_{c2}(T) - 4\pi M(H_{c2},T) \\ &\quad - \frac{0.022\alpha}{\lambda_{so}T_{c0}}[H_{c2}(T)+H_J(H_{c2},T)]^2 \\ &\quad - 3.56H^*_{c2}(0)\rho_{scat} \ , \end{aligned} \tag{2}$$

where $H^*_{c2}(T)$ is the orbital critical field coming from the interaction of the applied field with the conduction-electron orbits and $4\pi M(H_{c2},T)$ is the internal field arising from the alignment of the Gd^{3+} moments. The third term comes from the combined effects of the applied magnetic field and the exchange field H_J acting on the conduction-electron spins and is referred to as the paramagnetic limiting term; α is the Maki parameter and λ_{so} is the spin-orbit parameter. The effective field H_J is a first-order effect of the exchange interaction between the magnetic ions and the conduction electrons. The last pair-breaking term, which is a second-order effect of the exchange interaction, is due to conduction-electron scattering from uncorrelated spins. This equation can be simplified further by neglecting the magnetic field $4\pi M$, which reaches a maximum value of only ~ 120 Oe for $x=1.987\%$.[27] Also, the fact that $T_c(x)$ for

TABLE IV. Coefficients of the low-temperature specific-heat fits of the data in the T interval 13–22 K to $C=\gamma T+\beta T^3$ and the deduced Debye temperature Θ_D as well as the estimated superconducting specific-heat jump ΔC, superconducting transition temperature T_c, and electronic specific-heat coefficient γ (inferred from extrapolation of the data above T_c to 0 K).

x %	γ (mJ/mol K^2)	β (mJ/mol K^3)	Θ_D (K)	ΔC (mJ/mol K)	T_c[a] (K)	$\gamma(T=T_c)$ (mJ/mol K^2)
0	106.3	0.109	630	1606	0.88	890
0.228	73.7	0.388	412	1168	0.73	862
1.018	62.17	0.488	382	b	<0.5	800
1.987	43.96	0.500	379	b	<0.5	750

[a]Deduced from specific-heat measurements.

[b]The transition was not within the temperature range of our calorimeter.

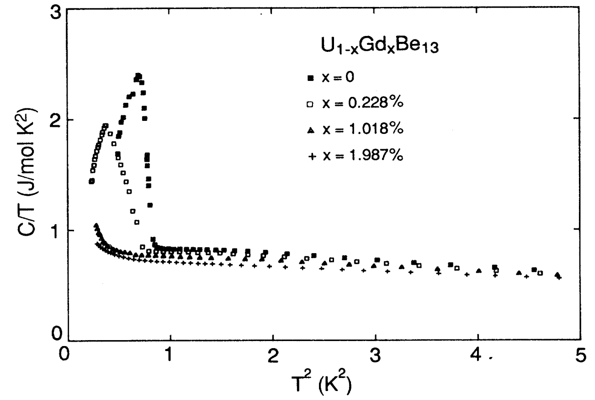

FIG. 9. Specific heat C divided by T vs T^2 data for $T^2 < 5$ K^2; the specific-heat jumps for $x = 0$ and 0.228 %, below 1 K, are transitions to the superconducting state.

$U_{1-x}Gd_xBe_{13}$ falls between the corresponding values of $T_c(x)$ for $U_{1-x}Lu_xBe_{13}$ and $U_{1-x}La_xBe_{13}$ (Ref. 28) indicates a very weak pair-breaking effect due to exchange scattering from uncorrelated Gd spins, which suggests that the term $3.56H^*_{c2}(0)\rho_{\rm scat}$ in Eq. (1) can also be neglected. The expression for $H_{c2}(T)$ then reduces to

$$H_{c2}(T) = H^*_{c2}(T) - \frac{0.022\alpha}{\lambda_{\rm so}T_{c0}}[H_{c2}(T) + H_J(H_{c2},T)]^2 . \quad (3)$$

Hence the primary mechanism for breaking superconducting electron pairs is the Zeeman interaction of the exchange field H_J, associated with the Gd spins, with the superconducting electron spins. The orbital critical field H^*_{c2} in the $U_{1-x}Gd_xBe_{13}$ system was determined in two steps: First, $H^*_{c2}(x=0,T)$ was chosen so that Eq. (3) of the pair-breaking theory describes the experimental $H_{c2}(T)$ data of pure UBe_{13}, after which it was scaled with T_c, i.e.,

$$H^*_{c2}(T) \equiv H^*_{c2}(x,T) = \frac{T_{c0}(x,H=0)}{T_{c0}(x=0,H=0)} H^*_{c2}(x=0,T) . \quad (4)$$

The same method has been applied to the $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$ system where the reference compound is $(U_{0.971}Th_{0.029})Be_{13}$. The calculated values of $H^*_{c2}(0)$ are in general higher than the experimental values of the critical field $H_{c2}(0)$. The Maki parameter α can be determined using the relation $\alpha = 7.62\times10^{-2}[H^*_{c2}(0)/T_c]$, and an estimate of the spin-orbit parameter $\lambda_{\rm so} = 2\hbar/3\pi\tau_{\rm so}k_BT_{c0} \approx 10$ was obtained from parameters calculated[1] within a free-electron model.

The exchange field H_J associated with the Gd spins is given by

$$H_J = c\mathcal{J}(g_J-1)\frac{\langle J\rangle}{2\mu_B} , \quad (5)$$

with $\langle J\rangle = JB_J(g_J\mu_BJH/k_BT)$. The parameters are defined as follows: $c = x/14$ is the concentration of paramagnetic ions per formula unit, g_J is the Landé g factor, J is the total angular momentum of the rare-earth ion's Hund's rules ground state, B_J is the Brillouin function, and $\mathcal{J}$ is the exchange interaction parameter which is treated as an adjustable parameter in our fittings. Using the parameter values $g_J = 2$, and $J = \frac{7}{2}$, Eq. (5) becomes

$$H_J\ (\text{kOe}) = 2.16\times10^4\mathcal{J}B_J(7\mu_BH/k_BT)x . \quad (6)$$

The calculated $H_{c2}(T)$ curves using Eq. (3) and the parameters listed in Table I are shown in Fig. 10 for $U_{1-x}Gd_xBe_{13}$ and $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$, and compared to the data. For $U_{1-x}Gd_xBe_{13}$, the calculated curves in Fig. 10(a) do not describe the measurements when $H_J = 0$ (solid line) and fall above the real data at all fields and temperatures. As x increases, the discrepancy increases even more. Within the limitations imposed by

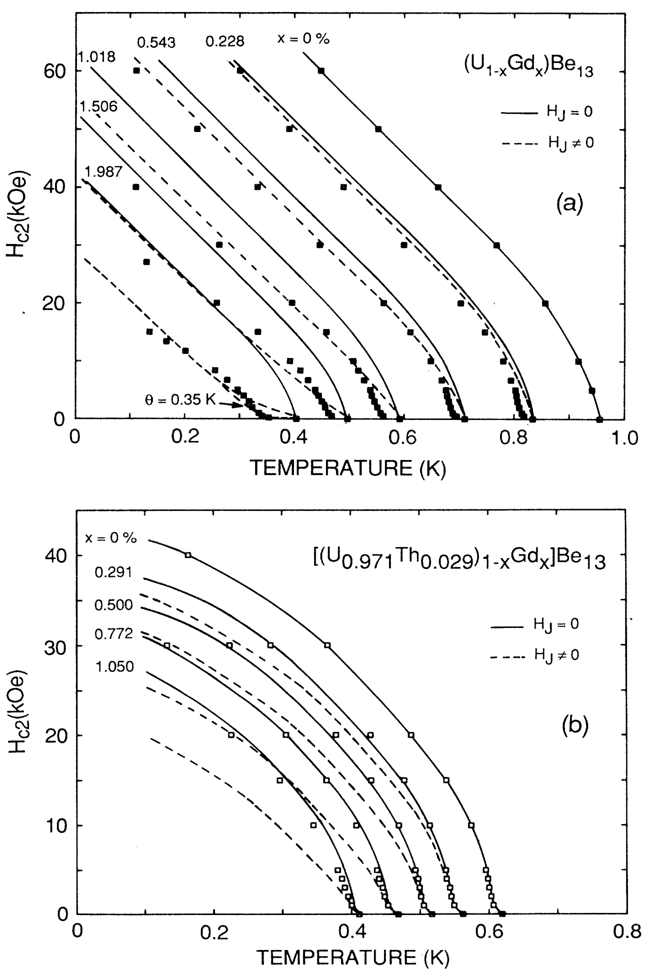

FIG. 10. Calculated curves of $H_{c2}(T)$ for (a) $U_{1-x}Gd_xBe_{13}$ compounds and (b) $(U_{0.971}Th_{0.029})_{1-x}$ Gd_xBe_{13} compounds using the multiple pair-breaking theory described in the text. The solid lines, for $H_J = 0$, and the dashed lines, for $H_J \neq 0$, are compared to the data.

our simplifying assumptions, the multiple pair-breaking theory with $H_J \neq 0$ seems to provide a satisfactory qualitative description of the $H_{c2}(T)$ data for Gd-doped UBe_{13} and the best results are obtained for $\mathcal{J}=0.036$ eV. However, the calculated curves do not describe the data in low fields very well, although the discrepancy is reduced by including a molecular-field constant (or Curie-Weiss temperature) Θ in the argument of the Brillouin function of Eq. (6), i.e.,

$$H_J\ (\mathrm{kOe}) = 777.4 B_J[7\mu_B H / k_B(T-\Theta)]x \ , \qquad (7)$$

as illustrated for the $x=1.987\%$ data when $\Theta=0.35$ K. For concentrations $x \leq 0.543\%$, the effect of Θ (not shown) is too small to make a difference so that the calculated curves for $\Theta=0$ and 0.8 K fall on top of each other. For $x=1.018\%$ and 1.506%, the low-field calculation is substantially improved by including values of Θ equal to 0.5 and 0.3 K, respectively. The low-field behavior of the $H_{c2}(T)$ data reflects, in our opinion, the depairing of superconducting electrons via the Zeeman interaction between the spins of the superconducting electrons and the exchange field H_J associated with the Gd^{3+} ions. As the Gd concentration increases and, in turn H_J, becomes nonnegligible compared to H_{c2} at low temperatures, the paramagnetic limitation arising from the pair-breaking effect of the magnetic field on the superconducting pairs[29,30] becomes very effective as evidenced by the bending of the $H_{c2}(T)$ curves of $U_{1-x}Gd_xBe_{13}$ especially for $x > 1\%$. A characteristic of singlet superconductors is that their upper critical field is limited[31] by

$$H_{\mathrm{limit}} = \frac{H_{c2}^{*} H_{p0}}{[2(H_{c2}^{*})^2 + H_{p0}^2]^{1/2}} \ , \qquad (8)$$

where H_{p0} is the paramagnetic limiting field. Therefore, the simplest interpretation of the $H_{c2}(T)$ measurements favors singlet spin pairing, perhaps of *d*-wave character, or Balian-Werthamer- (BW-) type triplet spin pairing, which has a superconducting excitation spectrum similar to that of an ordinary BCS superconductor.

In the Gd-doped $(U_{0.971}Th_{0.029})Be_{13}$ system, the calculated $H_{c2}(T)$ curves, shown in Fig. 10(b), are best described when $H_J=0$ (solid lines). If the exchange field is included (dashed lines), the calculated $H_{c2}(0)$ values, using the parameters listed in Table II, are smaller than the experimental values by as much as 30%. For *p*-wave pairing,[32] the paramagnetic limit does not apply because the paramagnetic susceptibility of the superconducting state is essentially the same as that of the normal state, similar to what was proposed by Anderson and Morel[8] for ^3He. Accordingly, the insensitivity of the upper critical field in $(U_{0.971}Th_{0.029})Be_{13}$ to the strength of the Gd exchange field as evidenced by the scaling behavior of the normalized $H_{c2}(T)/H_{c2}(0)$ data with T/T_c indicates that paramagnetic limitation does not apply in this case and that the results are consistent with an Anderson-Brinkman-Morel- (ABM-) type triplet spin pairing. The specific heat has been measured in UBe_{13} and found to fit a T^3 power law in the superconducting state at low temperatures. The power-law behavior is consistent with an energy gap vanishing at points on the Fermi surface, i.e., an ABM state.[33] However, the absence of paramagnetic limitation also occurs for singlet spin pairing or BW-type triplet spin pairing when the spin-orbit interaction is sufficiently strong to significantly increase the superconducting-state spin susceptibility and, in turn, the paramagnetic limiting field. The influence of the spin-orbit scattering parameter λ_{so} on the calculated $H_{c2}(T)$ is illustrated in Fig. 11 for $x=0.291$ and 0.772 %. It can be seen that unphysically large values of λ_{so}, as much as 500, would be required to effectively eliminate the paramagnetic effect in these materials.

From low-temperature specific-heat data[33] in UBe_{13} which shows a T^3 dependence below T_c, no firm conclusions as to the nature of the pairing can be drawn because a T^3 dependence in C is possible for both triplet and singlet spin pairing. The effect of impurities on the electronic specific-heat coefficient γ of UBe_{13}, which is a measure of correlations in the heavy-electron state, is less pronounced than on the critical temperature T_c. For $x=0.228\%$, T_c decreases by 17%, whereas γ is reduced by only 3%. On a $\Delta C/\Delta C_0$-versus-T_c/T_{c0} plot, where ΔC_0 and T_{c0} refer to $x=0$, the reduced specific-heat jump $\Delta C/\Delta C_0$ is equal to 0.73 for $x=0.228\%$ and does not fall on the BCS law of corresponding states line.[34] The value of $\Delta C/\Delta C_0$ is slightly smaller than the value of 0.76 calculated for pair breaking by long-lived local magnetic moments in the absence of the Kondo effect by Abrikosov and Gor'kov (AG).

The superconducting phase diagram in $U_{1-x}Th_xBe_{13}$ might involve more than the two superconducting phases probed in our experiments. In fact, Joynt, Rice, and Ueda proposed a phase diagram with three different anisotropic superconducting phases.[35] The transition at T_{c2} is apparently a second-order phase transition according to Ref. 17; Rauchschwalbe *et al.*[18] have argued that a second-order transition at T_{c2} implies that a new super-

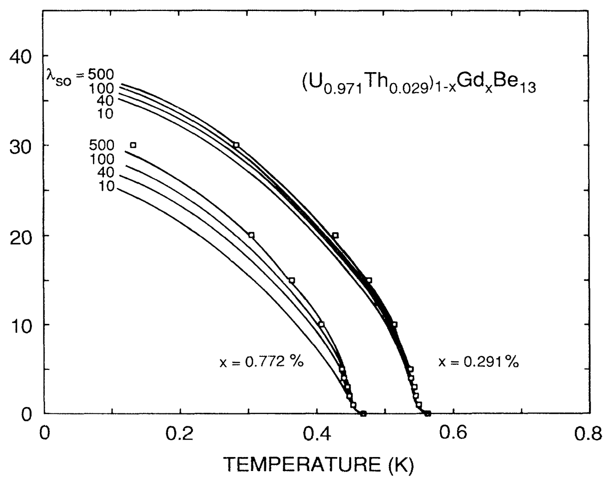

FIG. 11. Calculated curves of $H_{c2}(T)$ for two $(U_{0.971}Th_{0.029})_{1-x}Gd_xBe_{13}$ compounds ($x=0.291$ and 0.772 %) where the effect of spin-orbit interaction on the shape of these curves, when $H_J \neq 0$, is shown.

conducting order parameter forms below T_{c2} and coexists with the initial-order parameter formed below T_{c1}. The magnetic correlations observed for $x = 3.5\%$ can then be explained in terms of an unconventional superconducting state which belongs to a multidimensional symmetry group representation and hence can have spontaneous magnetic properties.[11] Kumar and Wolfle[36] have discussed a model for the coexistence of *s*- and *d*-wave superconductivity in $U_{1-x}Th_xBe_{13}$. They proposed that in pure UBe_{13} the high-temperature transition is to a *d*-wave state with a continuous evolution to the mixed *s*-*d*-wave state at lower temperatures, whereas in the Th-doped UBe_{13} the order is reversed and the second component (now of *d*-wave symmetry) is switched on via a second-order phase transition. In our experiments we were unable to probe the magnetic-field dependence of the superconducting transition at T_{c2}; however, such a study can be carried in a dilution refrigerator. Sigrist and Rice[37] have proposed a possible superconducting phase diagram for the $U_{1-x}Th_xBe_{13}$ system which reproduces the details of the $T_c(x)$ curve in zero applied field.

One interesting result that emerged from this work is illustrated in Fig. 12, where the change in the superconducting transition temperature $\delta T_c = T_{c0} - T_c$, in zero applied magnetic field, is plotted versus the rare earth R for $U_{0.985}R_{0.015}Be_{13}$ compounds (T_{c0} and T_c refer to pure and R-doped UBe_{13}, respectively). No data are shown for R = Pr, Pm, Eu, Tm, and Yb. In the context of the AG theory for paramagnetic impurities in conventional superconductors,[38] the decrease of T_c with paramagnetic impurity concentration should scale with the de Gennes factor $(g_J - 1)^2 J(J+1)$ of the R rare-earth ion. Instead, what is seen here is that for the heavy rare earths, T_c decreases monotonically and linearly with increasing atomic number of the substituents. The correlation of T_c with rare-earth atomic number is perhaps due to a volume effect corresponding to the UBe_{13} lattice contraction, as atoms smaller than U (specifically from Tb to Lu) are incorporated into the structure.[15,39] The existing data points when R atoms larger than U (specifically from La to Gd) are substituted seem to suggest that δT_c varies nonmonotonically in that region. The influence of pressure P on superconductivity in UBe_{13}, where a depression of T_c with increasing P was observed,[40] is consistent with the decrease of T_c when rare-earth atoms smaller than U are substituted. Interpretation of the data in Fig. 12 is complicated by the fact that both magnetic and nonmagnetic impurities have a depairing effect on the superconducting electrons in anisotropic *p*-wave[8] and *d*-wave[14] superconductors.

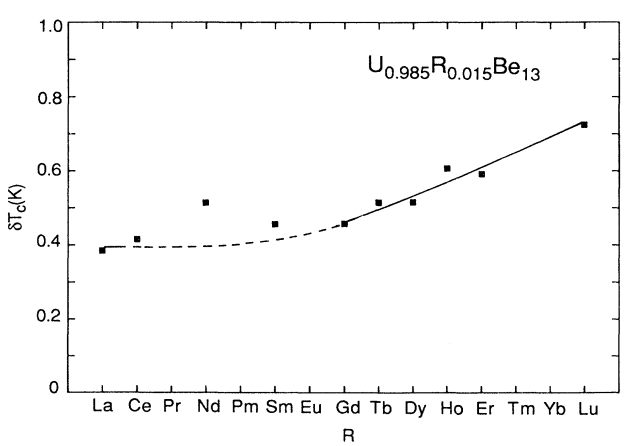

FIG. 12. Superconducting transition temperature change $\delta T_c = T_{c0} - T_c$ for some $U_{0.985}R_{0.015}Be_{13}$ compounds, where T_{c0} and T_c refer, respectively, to the pure and doped UBe_{13} compound.

CONCLUSIONS

The identification of the parity of the superconducting state and the classification of the symmetry of the superconducting order parameter in heavy-electron compounds is an important issue. Results of upper critical magnetic field $H_{c2}(T)$ measurements support the idea of different types of superconductivity in UBe_{13} and $(U_{0.971}Th_{0.029})Be_{13}$ as suggested by the pressure dependence of T_c in these systems. The compound UBe_{13} apparently exhibits singlet spin pairing or BW-type pairing, whereas $(U_{0.971}Th_{0.29})Be_{13}$ is in a triplet ABM-like state. The occurrence of at least two different superconducting states in $U_{1-x}Th_xBe_{13}$ is unique, so far, to Th impurities. Other nonmagnetic impurities do not produce the same effects as thorium, and the reason could be important to a proper understanding of the superconducting properties in UBe_{13}. From a theoretical point of view, it has been shown that antiferromagnetic fluctuations, which are observed in heavy-electron systems, assist even-parity pairing and impede odd-parity as well as isotropic even-parity pairing.[42] Considerations based on Monte Carlo simulations and on an Anderson lattice Hamiltonian also favor an anisotropic singlet superconducting state in the heavy-electron superconductors.[43] Several phenomenological theories have been advanced in order to explain the $T_c(x)$ phase diagram of $U_{1-x}Th_xBe_{13}$ which are based on (1) a multicomponent superconducting phase diagram,[35] (2) a splitting of the multidimensional representation of the superconducting state due to Th atoms disrupting the cubic symmetry,[44] (3) a crossing of two different types of anisotropic superconductivity at $x \approx 1.8\%$,[36,37,45] and (4) an interaction between superconductivity and coherent Kondo scattering where the lower transition in the specific heat is due to a transition to the coherent state.[24]

ACKNOWLEDGMENT

The research at University of California, San Diego (UCSD) was supported by the U.S. Department of Energy under Grant No. DE-FG03-86ER45230 (B.W.L, S.E.L., and M.B.M.) and the U.S. National Science Foundation–Low-Temperature Physics Grant No. DMR-87-21455 (Y.D. and M.B.M.). Research at Los Alamos National Laboratory (LANL) was carried out under the auspices of the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences.

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