Effects of boron substitution on the superconducting state of UBe_{13}

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Specific heat, magnetic susceptibility, and resistivity were measured on polycrystalline samples of $U(Be_{1-x}B_x)_{13}$ with $0 \le x \le 5.2 \times 10^{-3}$. Small amounts of B greatly affect the specific-heat discontinuity at the superconducting phase transition in UBe_{13} , while only weakly decreasing the superconducting transition temperature. The presence of B impurities also influences the resistivity. These observations are discussed in terms of possible impurity-induced changes in the symmetry and/or pairing interaction of the superconducting state in UBe_{13} .

I. INTRODUCTION

For almost a decade' there has been a great deal of interest in the exotic superconducting properties of UBe_{13} . Measurements² sensitive to thermal excitations of quasiparticles show power-law temperature dependencies, indicating that an anisotropic superconducting gap exists with either an axiallike or polarlike gap structure. Theoretical models incorporating superconducting order parameters with nonzero angular momentum components and possible nonsinglet spin states have been used to describe these properties.³

While investigations of the symmetry of the gap structure continue, the mechanism for the superconducting pairing interaction has also yet to be confirmed. The evidence for nodes in the superconducting gap structure mentioned above, the presence of antiferromagnetic (AFM) correlations in most heavy-fermion superconductors, and the strong on-site Coulomb repulsion between the superconducting f electrons has led to the inference that AFM spin fluctuations rather than phonons play an important role in mediating the attractive pairing interaction. Theoretical studies⁴ further confirm this concept, although no experimental proof for this hypothesis exists. For example, although neutron-scattering experiments⁵ have found evidence for AFM correlations in the normal state of many heavy-fermion superconductors, quasielastic neutron-scattering experiments in polycrystalline UBe₁₃ have not been able to confirm the AFM nature of the low-lying excitations.⁶ However, recent stud- ies^7 of the field dependence of the resistivity and specific heat do provide evidence for spin fluctuations below about 5 K in $(U, Th)Be_{13}$. In this paper we present additional data on B-doped UBe_{13} and discuss the implications of these data for the superconducting properties of this system.

It is well known that the ground-state properties of UBe_{13} are very sensitive to certain impurities. For example, small amounts of Th substituted for U in U_{1-v} Th_vBe₁₃ produce a nonmonotonic reduction of the superconducting transition temperature T_c , an increase in the linear coefficient of specific heat γ at T_c , and two specific-heat anomalies in the superconducting state between $y \approx 0.019$ and 0.042.^{8,9} Measurements of the muon-spin relaxation μ SR rate demonstrate that some type of small-moment quasistatic magnetism sets in at the lower transition.¹⁰ Recently, it has been shown that B substitution for Be in $U(Be_{1-x}B_x)_{13}$ produces an enhanced specific-heat jump ΔC at T_c for $x = 2.3 \times 10^{-7}$ Equivalently to the in $U(Be_{1-x}B_x)_{13}$ produces an enhanced specific-heat jump ΔC at T_c for $x=2.3\times10^{-3}$ compared to $x=0.^{1,11,12}$ The ΔC is accompanied by an increase in the normal state γ , as in the case of U,Th)Be₁₃; however, μ SR has revealed no quasistate magnetic correlations below 1 K for $x = 2.3 \times 10^{-3}$.¹² To further investigate this phenomenon we present results of magnetic-susceptibility, resistivity, and specific-heat measurements for B concentrations $x=0$, 0.0008, 0.0023, 0.0034, and 0.0052.

The effects of B substitution are seen in all three measurements; however, the most interesting result is the B concentration dependence of ΔC . With increasing B content, ΔC goes through a maximum when $x \sim 2.3 \times 10^{-3}$. The transition temperature T_c changes less than 15% with the maximum B doping, while γ increases as the B concentration is increased. Below, we discuss these findings in terms of possible changes in the symmetry of the superconducting state and a possible relationship between the superconducting parameters and the characteristic energy scale associated with the spin fluctuations in this system, which we characterize by a spinfluctuation temperature T_0 .

II. MEASUREMENTS AND RESULTS

Polycrystalline samples of $U(Be_{1-x}B_x)_{13}$ with $0 \le x \le 5.2 \times 10^{-3}$ were arc melted using standard techniques. The selected 8 concentrations range from 0 to 6.8% relative to the U, spanning the same concentration range as Th in $\mathbf{U}_{1-\nu}$ Th_{ν}Be₁₃, where two superconducting transitions occur. Relative to Be, however, the range of 8 concentrations is more than an order of magnitude smaller and should not change the band filling by more than 0.5% . Despite the slightly smaller covalent radius of 8, no observable changes in the cubic lattice parameters were detected in x-ray diffraction patterns. NMR experiments¹³ on the B and Be nuclei find that the measured B concentration is approximately equal to the nominal values, and that the B impurities tend to sit exclusively on the cubic lattice sites, i.e., the $m3$ sites (see discussion below).

Measurements of the magnetic susceptibility were performed with a superconducting quantum interference device magnetometer manufactured by Quantum Design. Data were taken from $T=350$ to 1.8 K in an applied field of 0.¹ T. The resistivity was obtained between room temperature and 1.4 K with a standard four-probe technique. A thermal-relaxation technique¹⁴ was employed to determine the specific heat as a function of temperature from $T = 0.32$ to 20 K for samples several milligrams in size. Corrections to the relaxation time due to the thermal contact between the sample and its platform reduced the measured heat capacity by \sim 16% at the lowest temperatures for the samples with the worst thermal contact. These corrections became insignificant at higher temperatures.

The reciprocal susceptibilities for the pure sample and the one with the highest concentration of boron display Curie-Weiss-like behavior at high temperatures, as seen in Fig. 1. Consistent with previously published results,⁹ the effective moment μ_{eff} for $x = 0$ is $\sim 3.41 \mu_B$. At the highest B concentration ($x = 5.2 \times 10^{-3}$), there is a $\sim 2\%$ increase in μ_{eff} , which is just resolvable with our experimental uncertainty. This increase, however, is less than the variation in reported values for the pure material. If the effect is intrinsic, the presence of 8 appears to slightly enhance the $5f$ localization.

The incorporation of B into UBe_{13} also influences the scattering process of the charge carriers. Illustrated in Fig. 2, the peak in the low-temperature resistivity at $T=2.5$ K in pure UBe₁₃ moves to lower temperatures with increasing B. When $x = 5.2 \times 10^{-3}$, this narrow anomaly is no longer apparent above our lowest temperature, and only the very broad maximum at $T \approx 19$ K remains. The magnitude of the resistivity for $x = 5.2 \times 10^{-3}$ is also decreased below that of UBe₁₃ at all

FIG. 1. The reciprocal dc susceptibility for pure and Bdoped UBe_{13} as a function of temperature.

FIG. 2. The temperature-dependent resistivity for pure and B-doped UBe_{13} , after normalizing the data to the value at $T = 250$ K.

temperatures measured. This behavior is very similar to what occurs with Th substitution.^{7,8}

In Fig. 3 the measured specific heat for two concentrations is displayed as C/T versus T. The data show a relatively unenhanced specific heat at $T \ge 10$ K that gradually increases with decreasing temperature below 10 K, where a many-body heavy-electron state, associated with spin fluctuations, θ begins to develop at a temperature $\approx T_0$ (defined more precisely below). When $T = T_c \approx 0.9$ K, the abrupt discontinuity in C/T signals a phase tran-

FIG. 3. The specific heat divided by temperature versus temperature on a semilog plot for UBe_{13} and the B-doped sample with the largest superconducting specific-heat discontinuity.

sition into the superconducting state. Notice that the many-body enhancement of C/T between T_c and \sim 5 K for the $x=2.3\times10^{-3}$ concentration occurs at a lower temperature than for the pure sample, and that the specific-heat jump at T_c increases significantly for the Bdoped sample.

We have investigated the possibility that the enhanced jump in specific heat at T_c with B doping might be spurious, due either to two separate transitions occurring at almost the same temperature, or a redistribution of entropy associated with spin fluctuations in the normal state. The first case is not likely, for the following reasons. First, even though the maximum specific-heat discontinuity, which occurs when $x = 2.3 \times 10^{-3}$, is nearly twice as large as in the pure compound, the entropy associated with the transition has only increased by \sim 7.4%, as shown in Fig. 4. Second, the linear coefficient of specific heat extrapolated to zero temperature, as described below and shown in Fig. 6, increases by only $\sim 2.4\%$ in going from $x = 0$ to 2.3×10^{-3} . Third, the width of the transition does not change appreciably for small 8 concentration uses not enange uppressively T_c is narrower for $x = 2.3 \times 10^{-3}$ than for $x = 0$. Finally, muon-spin relaxation experiments were performed on the $x = 2.3 \times 10^{-3}$ sample, 12 and no internal quasistatic magnetism was observed above the detection limit of $0.001\mu_B$ per U site, eliminating the possibility of a second magnetic phase transition, as seen for certain Th concentrations. Thus there is no evidence for two separate phase transitions

which are coincident in temperature.

Since the entropy is the temperature integral of C/T , examination of Fig. 3 shows that the enhanced specificheat jump at T_c in the $x = 2.3 \times 10^{-3}$ B-doped sample results from a redistribution of the entropy into the transition region from both above and below. Note that this redistribution does not arise from a simple movement to lower temperatures of the spin-fluctuation entropy associated with the peak at $T \approx T_0$ in the specific heat. We therefore conclude that the enhanced ΔC must be connected with a change in the properties of the superconducting state itself.

The systematics of these effects were investigated by measuring the specific heat around the transition for four different values of x. These data are displayed in Fig. 5 (solid circles). Clearly the specific-heat jump at T_c increases, goes through a maximum, and eventually declines with increasing 8 doping. Because the superconducting anomalies are broadened slightly (probably by disorder), an entropy-conserving construction was used to analyze the data in the vicinity of T_c . This is shown by the solid lines in Fig. 5. A crossover from the linear extrapolation above and below T_c was determined by balancing the areas between the data and the extrapolated curves. In this manner both ΔC and T_c were defined. The zero-temperature limit of C/T for the normal state,

FIG. 4. Temperature dependence of the entropy for pure and B-doped UBe_{13} , (a) at low temperature and (b) at high temperatures.

FIG. 5. The specific heat divided by temperature as a function of temperature for pure and B-doped UB e_{13} . The solid circles are the measured data, and the lines are balanced entropy constructions used to determine T_c and ΔC (see text).

denoted by $\gamma_N(0)$, was estimated by adjusting the intercept of a linear extrapolation for C/T from $T=T_c$ to $T=0$ until the entropy at T_c is the same as measured in the superconducting state. The characteristic energy scale T_0 was defined experimentally as the temperature where $\gamma(T)$ decreases to $\gamma_N(0)/2$.

The values of $\gamma_N(0)$ and T_0 are displayed as a function of B concentration in Fig. 6, where one sees a significant decline in T_0 with increasing B content. We were unable to determine T_0 for the $x=5.2\times10^{-3}$ sample because the temperature where γ would attain half its zerotemperature limit is below T_c . Using the values for $\gamma_N(0)$ determined above, we calculated $\Delta C/\gamma_N(0) T_c$ for each concentration [Fig. 6(a)]. Notice that this ratio has a value close to the BCS result for s-wave superconductivity (1.43) for pure UBe_{13} and then increases through a maximum as B is added to the material. The maximum $\Delta C / \gamma_N(0)T_c$ equals 3.76 and occurs when $x \sim 2.3 \times 10^{-3}$. Even though values approaching this have been observed in some $A-15$ compounds,¹⁵ the maximum ratio in B-doped UBe_{13} is the largest of any known material. Despite this dramatic change in $\Delta C / \gamma_N(0)T_c$, the superconducting transition temperature is changed only somewhat [Fig. 6(b)]. We note that previously published⁹ values of $\Delta C/\gamma_N(0)T_c$ for UBe₁₃ are around 1.5, which is close to our result. The emphasis here is not on the exact magnitude of $\Delta C / \gamma_N(0)T_c$, which is subject to some systematic errors, but rather on the trend with x

FIG. 6. (a) $\gamma_N(0)$ and $\Delta C / \gamma_N(0)T_c$, defined in the text, as a function of B concentration x in U(Be_{1-x}B_x)₁₃. (b) The B concentration dependence of the superconducting transition temperature T_c and the temperature that characterizes the renormalized heavy-mass state T_0 . The lines are guides to the eye. For the highest B concentration, T_0 could not be determined because it falls below T_c .

and on the large enhancement at $x = 2.3 \times 10^{-3}$.

We also note an interesting correlation between our results and specific-heat measurements under hydrostatic pressures on pure UBe_{13} .¹⁶ Increasing the pressure to 9.3 kbars increases T_0 (determined by our definition) and decreases ΔC . Again a corresponding change is observed between T_0 and ΔC , though the trend with pressure is opposite to that for B impurities.

III. DISCUSSION

Strong-coupling effects can cause $\Delta C / \gamma_N(0) T_c$ to deviate from the BCS prediction. Assuming the Eliashberg formalism is applicable to these materials, it is possible to derive 17 an expression for the free-energy difference in the superconducting state from the nonlinear Matsubara-gap equations and the Bardeen-Stephen free-energy formula, which includes strong-coupling corrections. By taking the second derivative of the free-energy difference with respect to temperature and evaluating it at T_c , an expression for $\Delta C/\gamma_N(0)T_c$ is found in terms of the strongcoupling parameter T_c/ω_0 . The frequency ω_0 characterizes the bosonic excitation mediating the pairing interaction. If this boson is a phonon, ω_0 is the Debye frequency in the weak-coupling reduction of the expression. For heavy fermions, the boson may be different.

Extensive calculations of the dependence of $\Delta C / \gamma_N(0) T_c$ on T_c / ω_0 have been carried out for an isotropic $(s$ -wave) gap structure.¹⁷ This work has been exended^{18,19} to include anisotropic $(d$ -wave) superconduc tors whose pairing is driven by antiferromagnetic spin fluctuations characterized by a δ -function spectral density at ω_0 . Although the parity of the superconducting state in UBe $_{13}$ has not yet been established, we shall use these two (even parity) model calculations as examples of effects on isotropic and anisotropic superconducting states to help elucidate our data. As T_c/ω_0 approaches zero, the weak-coupling limits for the two models are different. For s-wave pairing $\Delta C / \gamma_N(0)T_c = 1.43$ (the BCS value), while this ratio is always smaller for the d wave case.^{19,20} (The actual value of the *d*-wave ratio is model dependent, 20 depending on the symmetry of the pairing state and the assumed spectral density of the bosonic exchange. A theoretical value as small as 0.67 has been obtained.¹⁹) When T_c/ω_0 is increased, the magnitude of $\Delta C/\gamma_N(0)T_c$ for the s-wave system passes through a maximum near $T_c/\omega_0 \approx 0.2$ and falls below the BCS value for $T_c/\omega_0 \ge 0.7$.¹⁷ For a *d*-wave system, calcuations also show an increase in $\Delta C / \gamma_N(0)T_c$ as T_c / ω_0 ncreases to about 0.25; 'model *d*-wave calculations at values of T_c/ω_0 exceeding 0.25 are not known to us.

The dependence of the transition temperature on the spectral function that characterizes the boson excitations responsible for mediating the superconductivity is also different for the isotropic and anisotropic models. In swave superconductors enhancing the fluctuation spectrum always increases T_c for all values of T_c/ω_0 . However, for the d-wave case low-frequency spin fluctuations decrease T_c because of pair breaking, while the spin fluctuations at high frequencies enhance T_c .¹⁸ Therefore, a crossover regime in the parameter space of T_c/ω_0 exists in d-wave superconductors, where T_c is not affected by changes in the spectral function. Finally, the calculations for d -wave pairing¹⁸ also suggest that the specific heat falls off more rapidly below T_c as the pairing strength is increased.

In addition to the dynamical effects of spin fluctuations on the pairing interaction, elastic and inelastic scattering of electrons from impurities are also important. In particular, in s-wave superconductors only magnetic scattering centers are pair breakers, depressing T_c . By contrast, both magnetic and nonmagnetic scatterers reduce T_c in an anisotropic superconducting state.¹⁸ Moreover, the consequences depend on whether the scattering is elastic or inelastic. Inelastic scattering can theoretically enhance T_c for an s-wave state, while depressing T_c for an anisotropic superconductor.²¹

In comparing these theoretical models to our data, we note that the existing thermodynamic² and NMR relaxation rate²² data on UBe₁₃ and (U,Th)Be₁₃ strongly suggest an anisotropic superconducting gap structure. Subsequent discussions of our results will be in this context. To begin with, we focus on the increased magnitude of $\Delta C / \gamma_N(0) T_c$, coupled with the relatively small change in T_c and $\gamma_N(0)$ in going from $x = 0$ to $x = 2.3 \times 10^{-3}$ (Fig. 6). We note that if undoped UBe_{13} enters a *d*-wave pairing state, then the coupling $\Delta C/\gamma_N(0) T_c \approx 1.5$ is already strongly enhanced compared to the predicted weakstrongly enhanced compared to the predicted weak-
coupling value.^{18,19} The additional enhancement of $\Delta C / \gamma_N(0)T_c$ with B doping could be due to at least two possible causes: (1) an increase in the coupling parameter T_c/ω_0 , which corresponds to a decrease in ω_0 since T_c is almost constant, or (2) a change in the symmetry of the superconducting state, possibly driven by an inelastic impurity scattering mechanism in such a way as to leave T_c unchanged. The first case would be consistent with models of d-wave pairing where T_c changes very little; the more rapid decrease in C/T below T_c for the $x = 2.3 \times 10^{-3}$ sample (Fig. 3) further supports this interpretation. We note that a decreasing spin-fluctuation temperature T_0 with increasing B concentration is also observed in the normal state just above T_c . This indicates a possible correlation between T_0 and ω_0 .

A second possible reason for the increased enhancement of $\Delta C / \gamma_N(0)T_c$ for $x = 2.3 \times 10^{-3}$ is a change from one nearly degenerate superconducting-state representation (d wave) to another (s wave). In this case, a significant change in the coupling strength would not be inferred for the following reason. The measured $\Delta C / \gamma_N(0)T_c$ values for $x=0$ (≈ 1.5) and $x=2.3\times 10^{-3}$ (≈ 3.8) would have to be compared to *different* weakcoupling limits, 1.43 for $x=2.3\times10^{-3}$ (s wave) and a smaller value for $x = 0$ (d wave). In this case, the measured $\Delta C / \gamma_N(0)T_c$ would indicate strong coupling, but without a large change in going from $x=0$ to $x = 2.3 \times 10^{-3}$

We find additional evidence for a change in the superconducting properties of B-doped UBe_{13} when examining the temperature dependence of the entropy S (Fig. 4). The entropy was determined by setting $C/T = 0$ at zero

temperature and linearly extrapolating between this point and the lowest measured temperatures. We assume that any residual zero-temperature contribution to C/T in the superconducting state is negligible, as discussed further below. This has only been experimentally verified in the $x = 0$ and $x = 2.3 \times 10^{-3}$ materials, however, where measurements down to 50 mK were previously carried out.²³ Because $C = T dS/dT$, the specific-heat discontinuity at T_c represents a sudden change in the slope of the entropy around T_c . The larger discontinuity in ΔC seen when $x = 2.3 \times 10^{-3}$ signifies a more rapid dropoff of the entropy below T_c , which indicates that the excitations freeze out faster with decreasing temperature. This provides additional evidence that B doping may alter the size and/or nodal structure of an anisotropic gap. In this regard it is noteworthy that the temperature dependence of C below about 0.15 K changes from T^3 in UBe₁₃ to T^2
for $x = 2.3 \times 10^{-3}$,²³ also implying a change in nodal structure.

The falloff in the normalized specific-heat jump $\Delta C/\gamma_N(0)T_c$ for B concentrations greater than $x = 2.3 \times 10^{-3}$ could be due to a number of sources. It is clear that much of the decrease comes solely from an increase in $\gamma_N(0)$, which accounts for all of the decrease in $\Delta C / \gamma_N(0)T_c$ for $x=3.4\times10^{-3}$ and about half of the decrease for 5.2×10^{-3} . The additional decrease in ΔC for $x = 5.2 \times 10^{-3}$ may be because B eventually disorders the system, possibly leading to a decease in the integrated spectral density of coherent spin fluctuations responsible for the pairing interaction. The increase in $\gamma_N(0)$ for $x > 2.3 \times 10^{-3}$ could in part be related to the onset of a residual $\gamma_s(0)$ with increasing B impurities, similar to that found²⁴ in Th-doped UBe₁₃ for Th concentrations where a second specific-heat jump is observed. Here $\gamma_{s}(0)$ refers to a finite linear contribution to the specific heat in the superconducting state as the temperature approaches zero. In $(U, Th)B_{13} \gamma_s(0)$ becomes comparable to $\gamma_N(T=T_c)$. We note that our data do not extend to low enough temperatures to validate this hypothesis, though a linear extrapolation of C/T to $T=0$ leads to $\gamma_s(0) \ll \gamma_N(T_c)$ for all measured B concentrations.

It is also possible that the spin fluctuation spectrum is altered in a fundamental way by the introduction of impurities on either the U or Be sites. These impurities would break translational invariance and create Kondo "holes" in the background periodic lattice that can resonantly scatter electrons.²⁵ We note that the resistivity near ¹ K is actually reduced by the addition of either Th or B, indicating a reduction in incoherent scattering. Consequently, spin-fluctuation spectral weight could be moved to these "holes," producing spin entropy below T_c (as shown clearly in Fig. 4 for $x = 5.2 \times 10^{-3}$) and modifying the pairing interaction. This could change the nodal structure of the superconducting energy gap, the coupling strength, or both. The increase in $\gamma_s(0)$ with either Th or B doping might therefore represent the contribution from resonantly scattered Kondo "holes. "

In conclusion, we have measured the magnetic susceptibility, resistivity, and specific heat in boron-doped UBe_{13} . The most interesting consequence of our investigation is the observation of a systematic change in the specific-heat discontinuity at T_c with B concentration. Despite the rather dramatic variation in $\Delta C / \gamma_N(0) T_c$, the transition temperature varies only modestly with B concentration. These experiments, when qualitatively interpreted in terms of strong-coupling theories of anisotropic superconductivity, indicate that the presence of B changes either the energy scale and spectral density of the superconducting pairing interaction and/or the symmetry of the superconducting state. To date there is no adequate microscopic theory to explain how relatively

small amounts of substitutional impurities can cause these changes.

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