Title
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Journal
Physica B+C, 135(1-3)

ISSN
0378-4363

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Publication Date
1985-12-01

DOI
10.1016/0378-4363(85)90422-x

Physica 135B (1985) 14–21
North-Holland, Amsterdam

NUCLEAR MAGNETIC RESONANCE AND HEAVY-FERMION SUPERCONDUCTIVITY IN UBe_{13} AND RELATED SYSTEMS

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We report the use of the ^{9}Be nuclear magnetic resonance to study heavy-fermion superconductivity in the $U_{1-x}Th_xBe_{13}$ alloy system, $x = 0$ and 0.033. The nuclear spin–lattice relaxation rate $1/T_1$, which yields information on thermal excitations in the superconducting state, is found to vary more slowly the temperature than expected for a conventional BCS superconductor with a nonzero energy gap Δ. This indicates an enhanced density of excitations for low energies $E \ll \Delta$. At intermediate temperatures $1/T_1T^3 \approx$ const. for UBe_{13}, which is consistent with highly anisotropic pairing; the specific temperature dependence suggests lines of zeros of Δ on the Fermi surface. The relaxation data do not agree qualitatively with theories of superconducting pair breaking due to paramagnetic impurities. At low temperatures ($T < 0.2$ K), $1/T_1T$ is approximately constant in UBe_{13}, with a value which decreases with decreasing applied field. This behavior is probably not due to direct relaxation by paramagnetic impurities, for which the temperature dependence would be in the opposite sense. For $x = 0.033$ $1/T_1$ varies less rapidly than T^3. The additional relaxation may be due to filling in of the gap by Th-induced pair breaking. The ^{9}Be spectra give no indication of additional broadening due to vortex-lattice inhomogeneity or (for $x = 0033$) of magnetic ordering at a second transition temperature T_{c2} below the superconducting transition (at T_{c1}). Our results are compared to NMR studies of $CeCu_2Si_2$, and to other experiments in heavy-fermion superconductors.

1. Introduction

Cerium- and uranium-based intermetallic compounds have recently been discovered [1–3] in which very massive ($m_{eff} > 200m_e$) itinerant electrons, or "heavy fermions", are unambiguously involved in the superconducting state. These heavy-fermion superconductors exhibit extremely enhanced normal-state specific heats at low temperatures, with linear specific-heat coefficients $\gamma \equiv C/T$ hundreds of times greater than in ordinary transition metals. A clear signature of heavy-fermion superconductivity is a correspondingly large discontinuity in the specific heat at the superconducting transition temperature T_c, which implies that the heavy fermions themselves participate in the superconducting pairing [4]. The possibility has been raised [5] of unconventional Cooper pairing in these materials, with non-BCS orbital and spin symmetries and strong energy-gap anisotropy.

Furthermore an interesting "double transition"

[(a)] Supported by NSF grants DMR-8115543 and DMR-8413730, and by the U.C. Riverside Academic Senate Committee on Research.
[(b)] Supported by NSF grant DMR-8409390 and by the UCLA Academic Senate Committee on Research.
[(c)] Work performed under the auspices of the U.S. Department of Energy.
[(d)] Work supported by the Schweizerische Nationalfonds zur Fördering der Wissenschaftlichen Forschung.

has been observed in the specific heat of the pseudobinary alloy system $U_{1-x}Th_xBe_{13}$ for x of the order of a few per cent [6]. Superconductivity appears below the higher transition temperature T_{c1}, and persists below the second transition temperature T_{c2}. T_{c1} decreases with Th concentration, but there is an unusual plateau in $T_{c1}(x)$ in the region where the double transition is observed. No evidence for macroscopic phase separation has been found, and the second transition seems to be accompanied by strong anomalies in the ultrasonic sound velocity [7] and thermal expansion [8].

Nuclear magnetic resonance (NMR) techniques have been used to probe superconductivity since before the appearance of the BCS theory [9]. The utility of NMR lies in its sensitivity to the behavior of local microscopic magnetic fields, both static and dynamic, in condensed matter. We review briefly the kinds of information which can be obtained using NMR which are relevant to the study of heavy-fermion superconductors.

Nuclear spin-lattice relaxation, the process by which a nuclear spin population distribution attains thermal equilibrium which the lattice, is due to coupling between nuclear spins and low-lying thermal excitations. In metals the dominant mechanism for nuclear spin-lattice relaxation is often the so-called Korringa mechanism, in which conduction electrons are spin-flip scattered by the nuclear moments. Paramagnetic impurities in a metal can also couple to the nuclei, and impurity-spin fluctuations can be an important relaxation time T_1 or, equivalently, the relaxation rate $R \equiv 1/T_1$, is the principal quantity of interest.

NMR spectra reflect the distribution of local static fields, and can be used, for example, to map out the field distribution in the mixed-state Abrikosov vortex lattice [9]. Large NMR frequency shifts and line broadening accompany magnetic ordering transitions [11], and NMR can therefore be used to search for such transitions in superconductors. NMR spectra are split by the electrostatic interaction between nuclear quadrupole moments and crystalline electric field gradients. The latter depend on crystal structure, and are correspondingly altered by structural modifications or phase transitions.

The three heavy-fermion superconductors with the largest effective masses observed to date are $CeCu_2Si_2$ [1], UBe_{13} [2], and UPt_3 [3]. Kitaoka et al. [12] reported the first NMR studies of the superconducting state in $CeCu_2Si_2$, using zero-field ^{63}Cu nuclear quadrupole resonance (NQR). Subsequently ^{63}Cu NQR and NMR in $CeCu_2Si_2$ [13,14] and ^9Be NMR in UBe_{13} [15,16] were investigated. The ^{29}Si resonance is also available in $CeCu_2Si_2$. It is unlikely, however, that a ^{195}Pt resonance will be easily observable in UPt_3, because the estimated relaxation time T_1 at low temperatures is prohibitively short.

This paper reports the results of our NMR studies in the superconducting states of the heavy-fermion superconductors $U_{1-x}Th_xBe_{13}$, $x = 0$ and 0.033. (Data from the normal state will be discussed only as they affect interpretation of superconducting-state results.) Section 2 describes ^9Be spin–lattice relaxation in $U_{1-x}Th_xBe_{13}$, $x = 0$ and 0.033, and section 3 gives results obtained from ^9Be resonance spectra in these materials. We compare our data to other experimental results in these and other HFS systems in section 4, and section 5 summarizes our conclusions. Some of this work has been reported previously [15,16].

2. Spin–lattice relaxation in $U_{1-x}Th_xBe_{13}$

NMR experiments have been carried out on two specimens of UBe_{13} ($x = 0$). Sample no. 1 was ground to a coarse powder (0.5 mm grain size), and sample no. 2 was in the form of several small crystallites (5 mm size). A small-crystallite sample of $U_{1-x}Th_xBe_{13}$, $x = 0.033$, was also investigated. Superconducting transition temperatures T_c were measured using an ac inductance technique, and found to be 0.85 K and 0.75 K for UBe_{13} sample nos. 1 and 2, respectively, in the applied field $B = \mu_0 H \approx 1.56$ T used for most of the experiments. For the $x = 0.033$ sample $T_c(1.56\ \text{T})$ was found to be 0.55 K. Standard pulsed NMR techniques were used to obtain relaxation times and field-swept spectra.

Fig. 1 gives the temperature dependence of the ^9Be spin–lattice relaxation time T_1 in UBe_{13} (sample no. 1), in the form $1/T_1T$ vs. tempera-

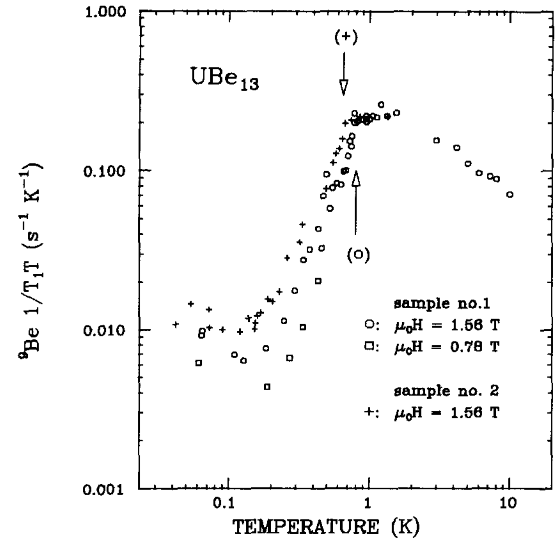

Fig. 1. Temperature and magnetic field dependence of the ^{9}Be spin-lattice relaxation time T_1, in the form $1/T_1T$ vs. temperature T. Transition temperatures in applied field are indicated by arrows. In the normal state and for $T < 0.1$ K $1/T_1T \approx$ const. The observed decrease of $1/T_1T$ with decreasing magnetic field would not be expected for relaxation by paramagnetic impurities.

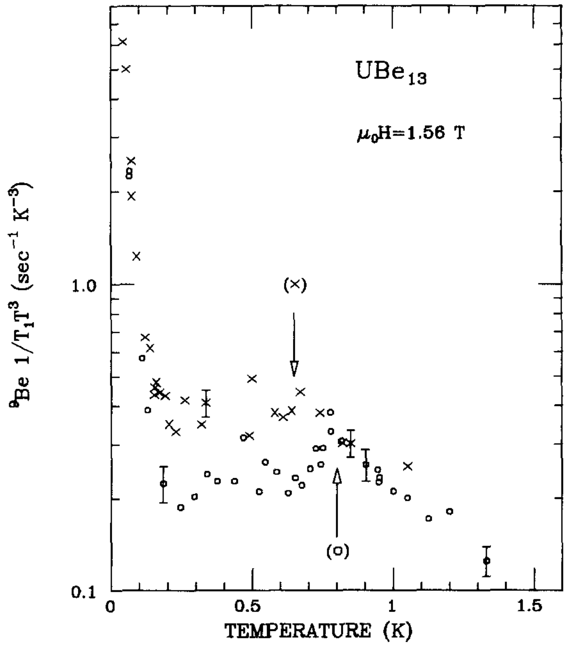

Fig. 2. Temperature dependence of the ^{9}Be $1/T_1T^3$ for two samples of UBe_{13}. Applied magnetic field $\mu_0H = 1.56$ T. Circles: sample no. 1. Crosses: sample no. 2. The arrows labeled by symbols give the superconducting transition temperatures $T_c(H)$ for each sample. Note the region $1/T_1T_3 \approx$ const. from ≈ 0.2 K to somewhat below $T_c(H)$.

ture, for several values of applied field. Korringa relaxation due to degenerate conduction-band electrons yields $1/T_{1n}T =$ const. in the normal state of a metal; this is confirmed in UBe_{13} for temperatures between T_c and about 2 K. Since $1/T_{1n} \propto T$, $1/T_{1s}T$ in the superconduting state is proportional to $T_{1n}(T)/T_{1s}(T) \equiv R_s(T)/R_n(T)$, where R_n is the relaxation rate which would be measured in the normal state at temperature T. R_s/R_n is directly available from theory [9], and $1/T_{1s}T$ is therefore a convenient quantity for comparison.

The salient features of these results are as follows: (1) The increase of $1/T_{1s}T$ just below T_c usually observed in conventional superconductors [9] is absent in UBe_{13}. (2) The data follow a power law $1/T_{1s}T \propto T^2$ between approximately 0.2 K and somewhat below T_c. This is more easily seen in fig. 2, which gives the temperature dependence of the quantity $1/T_1T^3$ for both $x = 0$ samples. (3) At temperatures below ≈ 0.2 K $1/T_{1s}T$ is again approximately constant, at a value ≈ 30 times smaller than in the normal state. We now discuss these features in turn.

The absence of a pronounced maximum in $1/T_{1s}T$ at intermediate temperatures and the power-law dependence of $1/T_{1s}T$ at intermediate temperatures are both consistent with marked broadening of the singularity at the gap parameter Δ in the BCS density of excited states [9]. The anisotropic form of the density of states obtained from the $L = 1$ (p-wave) triplet pairing model commonly used to describe superfluid ^{3}He [12, 17] provides such a broadening, although it is not unique in this regard.

The densities of states $N_s(E)$ for the so-called axial and polar anisotropic states are given in figs. 3a and 3b respectively. The BCS gap [N_s(BCS) = 0 for $E < \Delta$] is filled in by low-energy excitations ($E \ll \Delta_{max}$, where Δ_{max} is the maximum value of Δ). These come from excitations for which Δ nearly vanishes, i.e. near points (axial state) or lines (polar state) $\Delta = 0$ on the Fermi

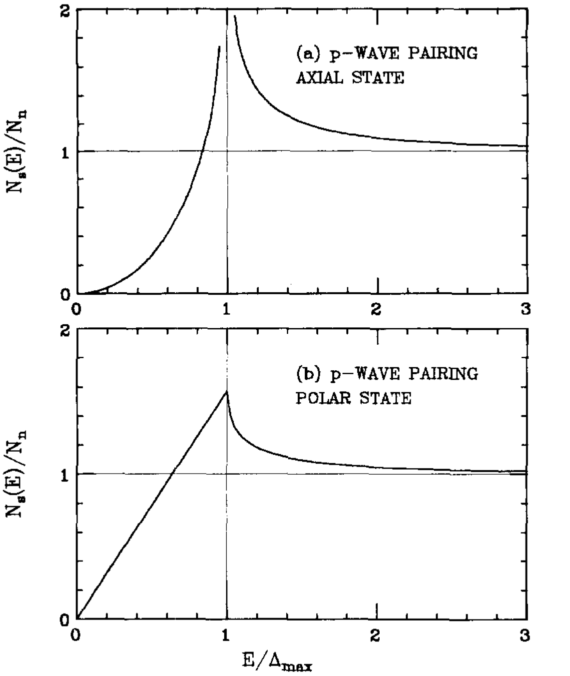

Fig. 3. Theoretical densities of superconducting quasiparticle excited states $N_s(e)$ for p-wave anisotropic superconducting pairing. (a) Axial (ABM) state [$N_s(E) \propto E^2$ for $E \ll \Delta_{max}$]. (b) Polar state [$N_s(E) \propto E$ for $E \ll \Delta_{max}$].

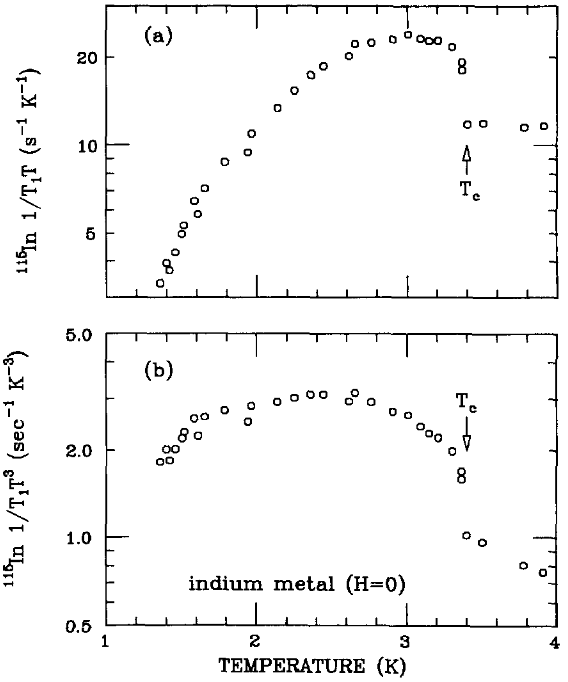

Fig. 4. ^{119}In spin–lattice relaxation in superconducting indium metal. (a) Temperature dependence of $1/T_1T$. (b) Temperature dependence of $1/T_1T^3$. The vertical axis of (b) has roughly the same scale as in fig. 2. Data from ref. 18.

surface. For these excitations $N_s(E) \propto E^q$, which leads to

$$R_s/R_n \propto T^p \quad (T \ll T_c), \qquad p = 2q\,. \tag{1}$$

for the temperature dependence of R_s/R_n at low temperatures. The values $p = 4$ and 2 are found for the axial and polar $L = 1$ triplet states, respectively. The latter is consistant with the data of fig. 2. This suggests that anisotropic pairing is a candidate for superconductivity in UBe_{13}. Low-lying excitations would then be associated, more generally than in the p-wave triplet model, with *lines* of zeros of Δ on the Fermi surface.

Fig. 4a gives the temperature dependence of the ^{119}In $1/T_{1s}T$ in indium metal, a conventional superconductor, for comparison with the UBe_{13} data of fig. 1 [18]. It can be seen that a pronounced maximum is observed just below T_c in indium, but not in UBe_{13} over the corresponding temperature range. Similarly, Fig. 4b gives the temperature dependence of $1/T_1T^3$ for indium metal. Downward curvature is observed which, if present, would also have been seen in UBe_{13} in spite of the poorer accuracy of the latter data (fig. 2). Indeed, the ^{115}In data can be fit very well [18] to the activated temperature dependence expected for a BCS superconductor with only slight anisotropy of the gap parameter [9], and the reasonable value $\langle\Delta\rangle_{av}/k_BT_c = 1.80 \pm 0.05$ is obtained [18]. This comparison indicates that UBe_{13} is a highly anisotropic superconductor compared to conventional materials.

The observation $1/T_{1s}T \approx$ const. below 0.2 K in UBe_{13} (fig. 1) suggests a nonzero $N_s(E)$ for $E = 0$, i.e. a small residual gaplessness. Other mechanisms exist for this observation, however, in particular pair breaking by paramagnetic spins (impurities or incompletely-compensated Kondo moments.)

The consequences for nuclear spin–lattice relaxation of the effect of pair breaking on excitations in a BCS superconductor have been treated in both the weak-coupling [19] and the strong coupling (Kondo) [20] limits. Pair breaking reduces the maximum value of R_s/R_n just below T_c, but theory does not yield the required linear density of states $N_s(E)$ at low energies. Fig. 5 gives the weak-coupling result for R_s/R_n vs T/T_c for several values of the pair-breaking parameter α required to suppress superconductivity completely $[T_c(\alpha_{cr}) = 0]$. It can be seen that quantitative agreement is lacking: a power law $R_s/R_n \propto T$ is found only for the value $\alpha \approx 0.91\alpha_{cr}$ which just suppresses the gap to zero, because in this case $N_s(E) \propto E^{1/2}$ [19].

Paramagnetic spins can also relax nuclei directly; indeed, this is more commonly observed than relaxation by superconducting excitations if the concentration of paramagnetic spins is at all large

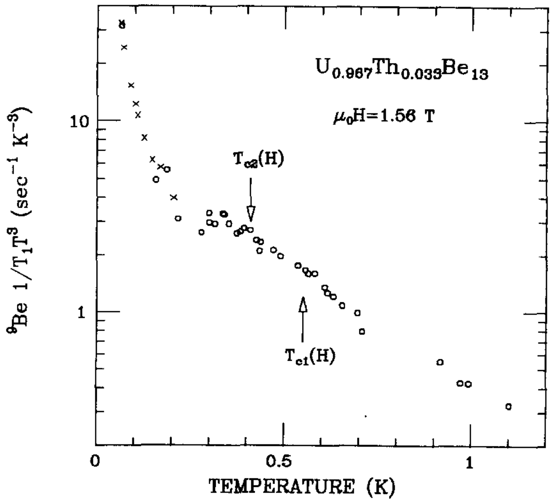

Fig. 6. Temperature dependence of the ^{9}Be $1/T_1T^3$ for $U_{1-x}Th_xBe_{13}$, $x = 0.033$. Circles and crosses give data taken on two separate runs. A law $1/T_1T^3 \approx$ const. is *not* observed below T_{c1}, in contrast to the result for $x = 0$ (fig. 2).

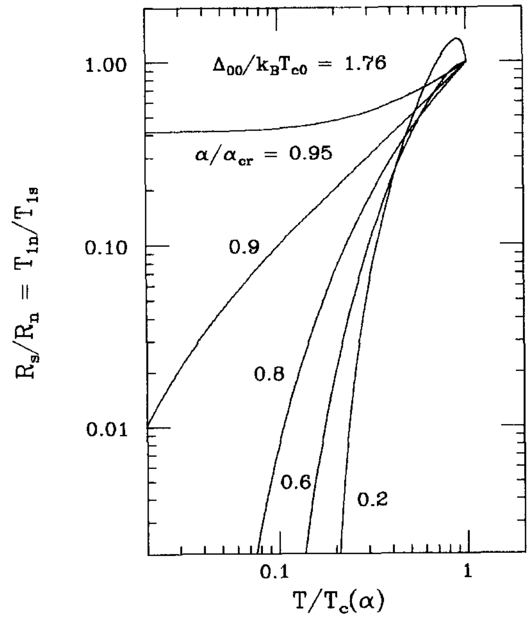

Fig. 5. Calculated temperature dependence of the normalized nucler spin–lattice relaxation rate R_s/R_n in a superconductor containing paramagnetic impurities, according to the weak-coupling theory (ref. 19). Here Δ_{00} and T_{c0} are the gap parameter and transition temperature in the undoped host superconductor, and α/α_{cr} is the normalized pair-breaking parameter.

[9]. However, in this case $1/T_{1s}T$ would be expected to be highly field dependent at low temperatures, due to saturation of the electronic spins by the field. Saturation decreases $1/T_1$ with increasing field [7], which is opposite to the observed effect. We conclude that direct paramagnetic relaxation is not likely to be an important contribution to the observed rate.

^{9}Be relaxation data from the $x = 0.033$ sample are given in fig. 6 in the form $1/T_1T^3$ vs. T. For $0.2\,K < T < T_c$, $1/T_1T$ varies less rapidly than T^2, as can be seen from the increase of $1/T_1T^3$ with decreasing temperature shown in Fig. 6. This result may indeed signal pair breaking due to Th impurities, which would depress T_c, as observed [21], and also broaden the structure in $N_s(E)$. The absence of structure in the neighborhood of either T_{c1} or T_{c2} should also be noted (fig. 6).

3. ^{9}Be resonance spectra in the superconducting state

NMR spectra were obtained from traces of integrated spin-echo intensity as a function of swept field. Spectra for UBe_{13} sample no. 2 are shown in fig. 7. These are typical quadrupole-split spectra for nuclear spin 3/2, with a central ($1/2 \leftrightarrow -1/2$) transition and quadrupole satellites. A curious property of these spectra is the absence of large shifts or broadening ΔH in the superconducting state; broadening ($\mu_0 \Delta H \approx 1$ mT) can be barely resolved at the lowest temperatures in the spectra of fig. 7. Slight broadening was previously found for the $x = 0.033$ sample [16], where a shift at $T = 0.113$ K was also observed. No shift is seen in Fig. 7. It is not clear how much of the previously-observed shift was due to field drift, however, since the field was regulated more closely for the $x = 0$ spectra than for $x = 0.033$.

A shift and broadening should arise from the inhomogeneous flux expulsion characteristic of the type-II mixed-state vortex lattice [9]. It is not clear why it sets in only for $T < 0.15$ K in UBe_{13}. The order of magnitude of the broadening, analyzed as described previously [16], leads to an estimated London penetration depth $\lambda_L > 2000$ Å and a corresponding Ginzburg–Landau parameter $\kappa > 20$ for $x = 0$ as well as 0.033.

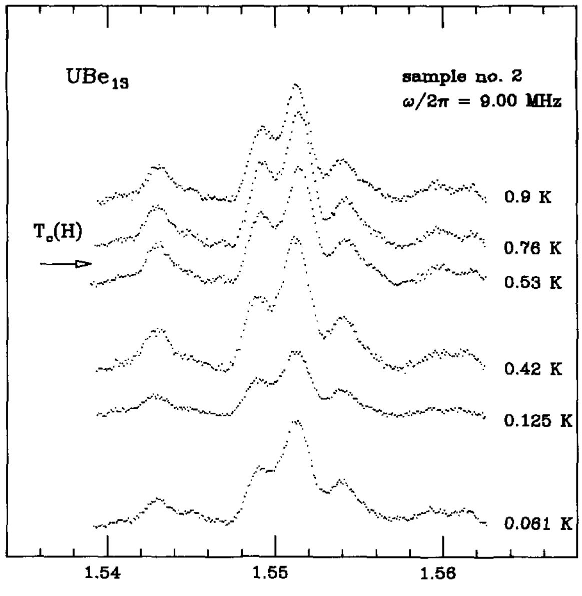

Fig. 7. Field-swept ^{9}Be spectra in the normal and superconducting states of UBe_{13} (sample no. 2). The shapes of the spectra are indistinguishable except at the lowest temperatures, where a small additional line broadening appears.

It has been remarked [16] that the absence of ^{9}Be shift or broadening at the second transition (temperature T_{c2}) for $x = 0.033$ is evidence for the absence of magnetic ordering below this temperature. For example, ^{27}Al NMR and NQR in the Kondo compound $CeAl_2$ [22], which orders antiferromagnetically at $T_N = 3.8$ K, exhibit broadened spectra ($\mu_0 \Delta H \approx 10$ mT) below T_N. NMR is a direct magnetic probe of local magnetic order, and it would be very hard to see how order with an appreciable magnetic moment could occur without causing NMR broadening. A quantitative estimate of this broadening is hard to make, since indirect (RKKY) couplings, etc., are not known in UBe_{13}. A rough estimate of the dipolar coupling is 0.1 T/μ_B [16], which leads to an upper limit of $\approx 10^{-2} \mu_B$ on the moment per U atom if the low-atom if the low-temperature broadening were magnetic in origin.

4. Comparison with other experiments

4.1. ^{63}Cu NQR and NMR in $CeCu_2Si_2$

NQR data show an absent [12] or strongly reduced [13] maximum in $1/T_1T$ below T_c, as in UBe_{13}, but the temperature dependence of $1/T_1T$ is much weaker below $\approx 0.6 T_c$ than in UBe_{13}. In an applied field of 0.572 T, however, the ^{63}Cu relaxation rate in $CeCu_2Si_2$ varies with temperature similarly to the ^{9}Be rate in UBe_{13} [14]. This strong-field dependence in $CeCu_2Si_2$ is opposite to that found in UBe_{13}, and is consistent with direct relaxation by paramagnetic impurities [14], possibly "remagnetized" Ce ions near defects. A direct mechanism was not favored in the original report [12], because the nuclear magnetization recovery was observed to be exponential [7]. A simple calculation shows, however, that spin-diffusion-limited exponential relaxation would be consistent with the data for impurity concentrations ≈ 1000 ppm.

4.2. Other techniques

The relative ultrasonic attenuation rate α_s/α_n

varies as T^2 at low temperatures in both UPt_3 [17] and UBe_{13} [23]. This is consistent with the nuclear spin–lattice relaxation results, except that the $1/T_1T \approx$ const. behavior seen in NMR in UBe_{13} below ≈ 0.1 K does not seem to be observed in ultrasonic attenuation. NMR relaxation might therefore be sensing a spin-dependent excitation which does not affect ultrasonic attenuation. Such a difference does not seem to arise from the present state of the theory, however.

The absence of strong NMR anomalies at either transition for the $x = 0.033$ sample makes it unlikely that the recently discovered giant ultrasonic anomalies at T_{c2} [7] can be attributed to an antiferromagnetic transition with an appreciable moment per U atom.

Thermal conductivity measurements in UBe_{13} [24] are also consistent with the picture which seems to emerge, at least at intermediate temperatures, of superconducting pairing with lines of zeros of the gap parameter.

5. Conclusions

NMR and data from other nonequilibrium techniques exhibit qualitative differences between heavy-fermion and conventional superconductors. The low-lying excitations are much more numerous in the former, and seem to be given by power-law dependences of the density of states on excitation energy. The power laws are consistent with excitation spectra obtained from ^{3}He-like theories of anisotropic pairing, but the experiments do not constitute proof of such pairing. For example, Kondo-lattice theories with singlet pairing are able to obtain lines of zeros of the gap parameter [25].

Surprisingly, ^{9}Be NMR spectra show no rapid onset of vortex inhomogeneity broadening below T_c in UBe_{13}. The same absence of anomalies in the NMR spectra are strong evidence against magnetic ordering at T_{c2} in $U_{1-x}Th_xBe_{13}$, $x = 0.033$. No anomaly was observed in the quadrupole splitting, either, but this only puts an upper bound of $\approx 2\%$ on any change of lattice parameter [16]. This would be an enormous structural change; a weaker one, consistent with the NMR spectra, could occur at the lower transition.

Acknowledgements

We are grateful for experimental help from K. Glover, J. Moore and C. Murayama. Thanks are due to P. Fulde, M.B. Maple, T.M. Rice, F. Steglich, M. Tachiki, C.M. Varma, and J.W. Wilkins for useful discussions.

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