LOW-TEMPERATURE $^9$Be SPIN RELAXATION IN SUPERCONDUCTING UBe$_{13}$

D E MacLAUGHLIN,* M D LAN,+ C TIEN,* J M MOORE,+W G CLARK,+ Z FISK,†
J L SMITH‡ and H R OTT∥

Department of Physics, University of California, Riverside, CA 92521, USA

The dependence of the $^9$Be spin-lattice relaxation rate $1/T_1$ on magnetic field has been measured in the heavy-fermion superconductor UBe$_{13}$ at temperatures well below $T_c$. A crossover between relaxation via spin diffusion to mixed-state vortex cores ($H > 6$ kOe) and to paramagnetic impurities ($H < 6$ kOe) is inferred.

1. Introduction

Nuclear spin-lattice relaxation studies of the heavy-fermion superconductors UBe$_{13}$ [1,2] and CeCu$_2$Si$_2$ [3] have yielded evidence for unconventional Cooper pairing in these exotic materials. In both systems the spin-lattice relaxation rate $1/T_1$ varies as $T^3$ over a considerable range of temperatures. In the case of UBe$_{13}$, however, the spin-lattice relaxation rate $1/T_1$ deviates from the $T^3$ law at lower temperatures, and varies as $T$ below $\approx 150$ mK [2]. It is obviously desirable to determine whether this deviation is extrinsic, e.g. due to paramagnetic impurities, or is an intrinsic feature of the superconducting state. We report in this paper field-cycling $^9$Be spin-lattice relaxation measurements in superconducting UBe$_{13}$ over a wide magnetic field range (20 Oe < $H < 15$ kOe) at two temperatures (67 and 147 mK), which were undertaken to clarify further the anomalous relaxation behavior described above.

2. Results

The field dependence of $1/T_1$ between 20 Oe and 16 kOe is given in fig. 1 at temperatures of 67 and 147 mK. For high fields ($H > 6$ kOe) $1/T_1$ approaches a linear variation in $H$, whereas at low fields the relation $1/T_1 \propto H^{-1/2}$ is an approximate fit to the data over more than two decades of field variation. Here it can also be seen that, at least for these two temperatures, $1/T_1$ varies essentially linearly with temperature at constant field. We now consider the origins of these features.

---

* Supported by US NSF grant no. DMR-8413730 and the UC Riverside Academic Senate Committee on Research
† Department of Physics and Solid Science Center, University of California, Los Angeles, CA 90024, USA. Supported by US NSF grant no. DMR-8409390 and the UCLA Academic Senate Committee on Research
‡ Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA. Work performed under the auspices of the US Department of Energy
§ Laboratorium fur Festkörperphysik, ETH-Honggerberg, CH-8093 Zurich, Switzerland. Supported by the Schweizerische Nationalfonds zur Förderung der Wissenschaftlichen Forschung

---

0304-8853/87/$03.50 \copyright$ Elsevier Science Publishers B V (North-Holland Physics Publishing Division)
2.1 High-field regime

Previous nuclear spin-lattice studies of conventional superconductors in the mixed state [4] revealed a breakdown of the activated behavior [5] expected for relaxation by quasiparticle excitations over the BCS energy gap. The excess relaxation rate varied linearly with both temperature and applied field, as in the present case. The mechanism suggested [4] for this breakdown invokes low-lying excitations [6] in Abrikosov vortex cores (of radius \( \approx \) the superconducting coherence length \( \xi \)), with energies similar to normal-state excitations, together with transfer of spin energy by spin diffusion between core and bulk nuclei. If spin diffusion is fast (\( DH/\Phi_0 > 1/T_1 \)), where \( D \) is the spin diffusion constant and \( \Phi_0 \) is the flux quantum, the order of magnitude of the observed spin-lattice rate is given by [7]

\[
1/T_1 = (H/\Phi_0) \xi^2/T_{1n} + [1-(H/\Phi_0)\xi^2]/T_{1s},
\]

where \( T_{1s} \) is the relaxation time due to superconducting excitations, i.e., far from cores. At low temperatures \( T_{1s} \) becomes very long, and the first term dominates.

This picture accounts for the low-temperature relaxation behavior in UBe\(_{13} \) at high fields (fig 1). The temperature and field dependences (\( 1/T_1 \propto HT \)) are consistent with the first term of eq (1), and the observed ratio (\( 1/T_1 \) obs)/(\( 1/T_1 \) n) \( \approx 25 \) at 15 kOe yields \( \xi = 350 \) Å at \( T/T_c \approx 0.1 \). This is in satisfactory agreement with the value of 140 Å derived from critical field measurements [8], considering the approximate nature of eq (1) and the unusual behavior of the critical field.

2.2 Low-field regime

Here the relaxation is clearly dominated by a different mechanism. We consider as a candidate for this mechanism relaxation via nuclear spin diffusion to dilute paramagnetic impurities. These are postulated to be present at some low concentration, too low to cause appreciable pair breaking [9] or other perturbation of the superconductivity. The impurities will, however, couple to nuclei via dipolar or indirect hyperfine interactions [10,11]. All these mechanisms yield a direct relaxation rate \( 1/T_1(r) \) of a nucleus a distance \( r \) from an impurity at the origin which is given, in the absence of spin diffusion, by

\[
1/T_1(r) = K/r^6,
\]

after angular dependences, RKKY sinusoidal variations (cos 2k_{	ext{pi}}r), etc., have been averaged over McHenry et al [11] have reviewed these coupling mechanisms, which involve either longitudinal or transverse fluctuations of the impurity electron spin. The impurity spin polarization \( B_j(x) \) and the transverse and longitudinal correlation times \( \tau_{\text{tr}}, \tau_{\text{L}} \) of the fluctuations enter in the form [11]

\[
K \propto \left( \frac{B_j(x)/x}{1 + (\gamma_1 \tau_{\text{tr}})^2 H^2} \right) \quad \text{(transverse fluctuations)}
\]

\[
K \propto \left( \frac{\text{d} B_j/\text{d} x}{1 + (\gamma_1 \tau_{\text{L}})^2 H^2} \right) \quad \text{(longitudinal fluctuations)},
\]

depending on the mechanism. Here \( \gamma_1 \) and \( \gamma_l \) are the impurity and nuclear gyromagnetic ratios, respectively, and \( B_j(x) \), \( x = g_\mu_B H/k_{\text{B}} T \), is the Brillouin function appropriate to the impurity moment. It is likely that longitudinal fluctuations will dominate the relaxation, since \( \gamma_1 > \gamma_l \).

According to this model several relaxation regimes can be distinguished, depending on the relative strengths of \( K \) and the nuclear spin diffusion constant \( D \) [10]. One of these, the so-called diffusion-limited regime, yields a relaxation rate

\[
(1/T_1)_{\text{dl}} = (4 \pi/3) N c D^{3/4} K^{1/4},
\]

where \( N \) is the density of impurity sites per unit volume and \( c \) is the impurity concentration. We can therefore account for the low-field relaxation field dependence \( 1/T_1 \propto H^{-1/2} \) if (a) the impurity relaxation is in the diffusion-limited regime, and (b) the longitudinal impurity correlation time \( \tau_{\text{L}} \) is long, so that \( \gamma_1 \tau_{\text{L}} \gg 1 \).

2.3 Temperature dependence

The observed temperature dependence in the high-field regime follows naturally from the
normal-like quasiparticle excitations in vortex cores which, as in the normal state, give rise to a linear temperature dependence of the relaxation rate \( (1/T_1)_n \propto T \).

The temperature dependence in the low-field regime, on the other hand, must arise from a temperature dependence of the longitudinal impurity-spin fluctuation rate \( 1/\tau_{cl} \), \( D \) and all other factors in \( K \) are temperature independent for low fields. If \( 1/\tau_{cl} \) is due to relaxation by bulk superconducting quasiparticle excitations, then it might obey the same power law as the nuclear relaxation rate \( 1/T_1 \) at higher temperature \( 1/\tau_{cl} \propto T^3 \). (We note, however, that ESR measurements in the normal state of UBe\(_{13}\) doped with 4f paramagnetic impurities [12] do not yield the linewidth enhancement expected from relaxation by heavy electrons. In the slow-fluctuation limit \( K \propto 1/\tau_{cl} \), and therefore \( (1/T_1)_d \propto T^{3/4} \). This would not be distinguishable from a linear temperature dependence in the data of fig 1.

2.4 Crossover from low to high field

The observed relaxation rate should then be the sum of eq (1) [with negligible \( 1/T_n \)] and eq (4). This is of the form

\[ 1/T_1 = AH^{-1/2} + BH, \]

(5)

if there is no field dependence other than that discussed above. If matched to the low- and high-field data of fig 1, eq (5) lies above the data in the crossover region. The dependence of eqs (3) on \( B_j(x) \) cannot be neglected, however, at the low temperatures of these measurements \( \mu_B H \) and \( k_B T \) are roughly equal in the vicinity of the field indicated by the arrow in fig 1. Above this field the impurity spins are saturated, and both \( B_j(x) \) and \( dB_j/dx \) decrease [as \( 1/x \) and \( \exp(-2x) \), respectively]. Such a decrease would rapidly remove the diffusion-limited component of the observed relaxation.

3. Conclusion

We have found an unexpected nonmonotonic dependence of the \(^9\)Be spin-lattice relaxation rate on applied field in UBe\(_{13}\), well below the superconducting transition temperature. The most conventional explanation ascribes the high-field regime to relaxation by spin diffusion to vortex cores, and the low-field relaxation is attributed to spin diffusion to paramagnetic impurities in a particular (diffusion-limited) relaxation regime. Other speculative features, such as a second band of (light) nonsuperconducting electrons, an excess of low-lying quasiparticle excitations, or a line of phase transitions at \( \approx 6 \) kOe, do not seem to be required.

The authors are grateful to C T Murayama for help with the low-temperature measurements.

References

[1] D E MacLaughlin, C Tien, W G Clark, M D Lan, Z Fisk, J L Smith and H R Ott, Phys Rev Lett 53 (1984) 1833
[2] C Tien, D E MacLaughlin, M D Lan, W G Clark, Z Fisk, J L Smith and H R Ott, Physica 135B (1985) 14
[3] Y Kitaoka, K Ueda, T Kohara, K Asayama, Y Onuki and T Komatsubara, J Magn Magn Mat 52 (1985) 341
[4] B G Silbernagel, M Weger and J E Wernick, Phys Rev Lett 17 (1966) 384
[5] L C Hebel and C P Slichter, Phys Rev 113 (1959) 1504
[6] C Caroli, P G de Gennes, and J Matricon, Phys Lett 9 (1964) 307
[7] J B Goldberg and M Weger, J Phys Soc Japan 24 (1968) 1279
[8] M B Maple, J W Chen, S E Lambert, Z Fisk, J L Smith, H R Ott, J S Brooks and M J Naughton, Phys Rev Lett 54 (1985) 477
[9] A A Abrikosov and L P Gor’kov, Sov Phys JETP 12 (1961) 1243
[10] I J Lowe and D Tse, Phys Rev 166 (1968) 279
[11] M R McHenry, B G Silbernagel and J H Wernick, Phys Rev B 5 (1972) 2958, also M R McHenry, Ph D thesis, University of California, Santa Barbara (1971), unpublished
[12] F Gandra, S Schultz, S B Oseroff, Z Fisk and J L Smith, Phys Rev Lett 55 (1985) 2719