J. Phys.: Condens. Matter 15 (2003) S2043-S2046

PII: S0953-8984(03)62795-5

Unconventional superconductivity in ferromagnetic UGe₂: a ⁷³Ge nuclear magnetic resonance/nuclear quadrupole resonance study

H Kotegawa¹, S Kawasaki¹, A Harada¹, Y Kawasaki¹, K Okamoto¹, G-q Zheng¹, Y Kitaoka¹, E Yamamoto², Y Haga², Y Ōnuki^{2,3}, K M Itoh⁴ and E E Haller⁵

¹ Department of Physical Science, Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan

² Advanced Science Research Centre, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan

³ Department of Science, Graduate School of Science, Osaka University, Toyonaka, Osaka 560-8531, Japan

⁴ Department of Applied Physics and Physico-Informatics, Keio University, Yokohama 223-8522, Japan

⁵ Department of Materials Science and Engineering, University of California at Berkeley and Lawrence, Berkeley National Laboratory, Berkeley, CA 94720, USA

Received 12 November 2002 Published 4 July 2003 Online at stacks.iop.org/JPhysCM/15/S2043

Abstract

We report ⁷³Ge nuclear magnetic resonance/nuclear quadrupole resonance (NMR/NQR) measurements on the itinerant ferromagnetic superconductor UGe₂ at ambient pressure (P = 0) and P = 1.3 GPa. Measurements of the nuclear spin–lattice relaxation rate $1/T_1$ of the ⁷³Ge NMR at P = 0 have revealed a T_1T = constant behaviour well below $T_{Curie} = 52$ K, evidencing the presence of a non-zero density of states at the Fermi level for the upand down-spin bands in the ferromagnetic state. At P = 1.3 GPa, where the ferromagnetic transition is reduced to $T_{Curie} = 26$ K, the dependence on temperature (T) of NQR $1/T_1$ at zero field has demonstrated the onset of the superconducting transition at $T_c = 0.55$ K. The lack of a coherence peak in $(1/T_1)$ just below T_c , followed by a T^3 -like behaviour, provide compelling evidence for the unconventional nature of the superconducting state that coexists with the ferromagnetic state on a microscopic scale in UGe₂.

1. Introduction

Superconductivity (SC) has been discovered in the ferromagnetic state below the Curie temperature T_{Curie} in UGe₂ [1, 2], ZrZn₂ [3], and URhGe [4]. The coexistence of SC and ferromagnetism (FM) has attracted a great deal of interest, because SC and FM are generally

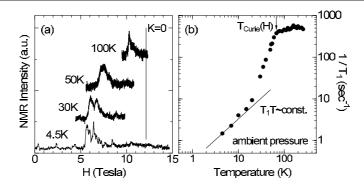


Figure 1. (a) The temperature dependence of NMR spectrum at ambient pressure (P = 0). The line at K = 0 indicates the magnetic field $H = 2\pi f_{NMR}/^{73}\gamma$ where f_{NMR} is the NMR frequency and $^{73}\gamma$ the gyromagnetic ratio of 73 Ge nuclei. (b) The *T*-dependence of $1/T_1$, which shows a sharp decrease below $T_{Curie}(H)$, followed by $T_1T \sim \text{constant below } \sim 20 \text{ K}$.

believed to be mutually exclusive. UGe₂ is an itinerant ferromagnet at $T_{Curie} = 52$ K at ambient pressure (P = 0), with a magnetic moment of 1.4 μ_B /uranium (U) atom [2, 5]. T_{Curie} decreases monotonically with increasing P. The SC sets in at pressures exceeding P =1.0 GPa, revealing a maximum value of $T_c \sim 0.7$ K even though the ferromagnetic moment remains large with 1.0 μ_B/U [2, 5]. Since the SC and FM are suppressed simultaneously at $P \sim 1.6$ GPa, the SC is considered to be relevant to an unexpected pairing mechanism inherent in the ferromagnetic state of this compound. An interesting point to address is what type of superconducting order parameter (OP) is realized in such an unconventional situation.

It is now quite controversial whether or not the SC and FM coexist at the microscopic level. It has been reported that the SC and FM are inhomogeneously separated and compete with each other [6]. Nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) are powerful tools enabling us to investigate the characteristics of both SC and FM from the microscopic point of view.

2. Experimental results

A polycrystalline sample with enriched ⁷³Ge was prepared and crushed into powder for NMR/NQR measurements. Figures 1(a) and (b) show the dependence on temperature (*T*) of the NMR spectrum and of $1/T_1$ for ⁷³Ge at P = 0, respectively. Since UGe₂ has a strong uniaxial magnetic anisotropy, the sample is expected to be oriented along the easy axis, that is, the *a*-axis [7].

While UGe₂ has three inequivalent Ge sites [8], a single peak in the NMR spectrum was observed for T higher than T_{Curie} , whereas it is roughly split into two peaks below T_{Curie} . The large negative shift from K = 0 is due to the development of ferromagnetic polarization under H as T decreases below T_{Curie} . The T₁-data plotted in figure 1(b) were measured at the peak on the low-field side in the spectrum. $1/T_1$ decreases below T_{Curie} , which is followed by T_1T = constant below ~20 K. This value of $1/T_1T$ should be related to the product $(N_{\uparrow}(E_F)N_{\downarrow}(E_F))$ of the respective densities of states $N_{\uparrow}(E_F)$ and $N_{\downarrow}(E_F)$ at the Fermi level for up- and down-spin bands [9].

Next, we present the remarkable results at P = 1.3 GPa, where the SC takes place at $T_c = 0.55$ K, as was confirmed by ac susceptibility data along with $T_{Curie} = 26$ K. Figure 2 shows the *T*-dependence of $1/T_1$ at P = 1.3 GPa and H = 0 that was measured

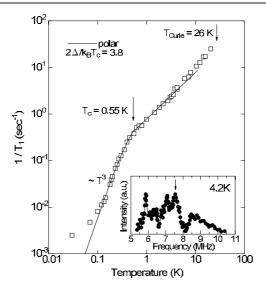


Figure 2. The *T*-dependence of $1/T_1$ at P = 1.3 GPa. The absence of a coherence peak just below T_c suggests unconventional SC in UGe₂. The solid curve was calculated by assuming a polar-type superconducting gap of $2\Delta/k_BT_c$ where the gap vanishes along a line. The inset shows the spectrum measured at H = 0. $1/T_1$ was measured at the peak indicated by the arrow.

at the peak denoted by arrow in the NQR/NMR spectrum indicated in the inset. The peak around ~7.6 MHz is observed on passing through T_{Curie} , suggesting that the NQR spectrum is somewhat affected by the appearance of an internal field at the three Ge sites induced by the spontaneous U-derived ferromagnetic moment. As shown in the figure, the $1/T_1T$ = constant relation is valid from T_c (=0.55 K) up to 5 K.

In the SC state, it is remarkable that $1/T_1$ decreases without any coherence peak just below T_c , this being followed by T^3 -like behaviour. These results provide microscopic evidence for bulk superconducting behaviour. The SC in UGe₂ is anticipated to belong to an unconventional class with a line-node gap, as other HF superconductors do. If we assume a polar p-wave model with a line-node gap, the magnitude of the SC gap is estimated to be $2\Delta/k_BT_c \sim 3.8$, as indicated by the solid line. The observation of T_1T = constant-like behaviour below 100 mK, however, prevents us from determining any precise gap structure. A future issue is hence whether the T_1T = constant-like behaviour below 100 mK arises from some impurity effect or a non-unitary odd-parity superconducting state [10, 11]. In any case, we emphasize that the NMR/NQR experiments have revealed the microscopic coexistence of SC and FM, because the *T*-dependence of $7^3(1/T_1)$ probes both anomalies inherent in the FM and SC.

3. Summary

We have reported the *T*-dependence of $1/T_1$ via ⁷³Ge NMR/NQR measurements on the ferromagnetic superconductor UGe₂ under P = 1.3 GPa. $1/T_1T =$ constant behaviour is observed well below T_{Curie} , whereas it is enhanced as the temperature approaches T_{Curie} . The bulk nature of the SC in UGe₂ was confirmed by the decrease in $1/T_1$ below $T_c = 0.55$ K without a coherence peak. It is possible that this *T*-dependence may be fitted by a polar p-wave model with a line-node gap. At the moment, however, the observation of $T_1T =$ constant-like behaviour below 100 mK does not allow us to propose any promising model for the SC in UGe₂.

An important outcome is that the respective anomalies relevant to both the FM and SC appear in the *T*-dependence of $1/T_1$ at T_{Curie} and T_c , and hence they appear to coexist on a microscopic scale in UGe₂.

Acknowledgments

The authors thank K Machida, N Tateiwa and J Flouquet for helpful discussions. This work was supported by the COE research grant (10CE2004) from MEXT of Japan. One of authors (HK) was supported by *JSPS Research Fellowships for Young Scientists*.

References

- [1] Saxena S S et al 2000 Nature 406 587
- [2] Huxley A et al 2001 Phys. Rev. B 63 144519
- [3] Pfleiderer C et al 2001 Nature 412 58
- [4] Aoki D et al 2001 Nature 413 613
- [5] Tateiwa N et al 2001 J. Phys. Soc. Japan 70 2876
- [6] Motoyama G et al 2001 Phys. Rev. B 65 020510(R)
- [7] Õnuki Y et al 1992 J. Phys. Soc. Japan 61 293
- [8] Kernavanois N et al 2001 Phys. Rev. B 64 174509
- [9] Ōnuki Y et al 1991 J. Phys. Soc. Japan 60 2127
- [10] Machida K et al 2001 Phys. Rev. Lett. 86 850
- [11] Fomin I A 2001 JETP Lett. 74 111

S2046