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# Unconventional superconductivity in ferromagnetic UGe<sub>2</sub>: a <sup>73</sup>Ge nuclear magnetic resonance/nuclear quadrupole resonance study

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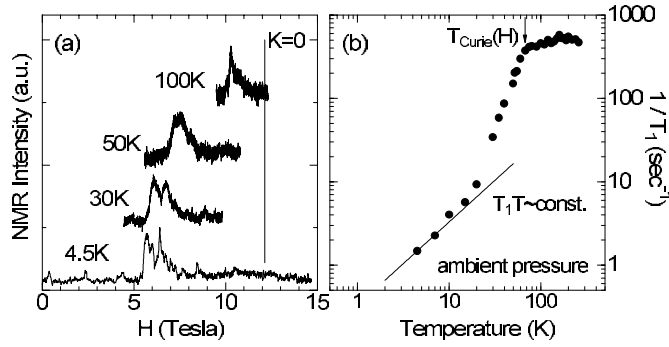
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## Abstract

We report <sup>73</sup>Ge nuclear magnetic resonance/nuclear quadrupole resonance (NMR/NQR) measurements on the itinerant ferromagnetic superconductor UGe<sub>2</sub> at ambient pressure ( $P = 0$ ) and  $P = 1.3$  GPa. Measurements of the nuclear spin–lattice relaxation rate  $1/T_1$  of the <sup>73</sup>Ge NMR at  $P = 0$  have revealed a  $T_1T = \text{constant}$  behaviour well below  $T_{\text{Curie}} = 52$  K, evidencing the presence of a non-zero density of states at the Fermi level for the up- and down-spin bands in the ferromagnetic state. At  $P = 1.3$  GPa, where the ferromagnetic transition is reduced to  $T_{\text{Curie}} = 26$  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 UGe<sub>2</sub>.

## 1. Introduction

Superconductivity (SC) has been discovered in the ferromagnetic state below the Curie temperature  $T_{\text{Curie}}$  in UGe<sub>2</sub> [1, 2], ZrZn<sub>2</sub> [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  $H = 2\pi f_{NMR}/\gamma$  where  $f_{NMR}$  is the NMR frequency and  $\gamma$  the gyromagnetic ratio of  $^{73}\text{Ge}$  nuclei. (b) The  $T$ -dependence of  $1/T_1$ , which shows a sharp decrease below  $T_{Curie}(H)$ , followed by  $T_1T \sim \text{const.}$  below  $\sim 20$  K.

believed to be mutually exclusive.  $\text{UGe}_2$  is an itinerant ferromagnet at  $T_{Curie} = 52$  K at ambient pressure ( $P = 0$ ), with a magnetic moment of  $1.4 \mu_B/\text{uranium (U)}$  [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  $1.0 \mu_B/\text{U}$  [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.

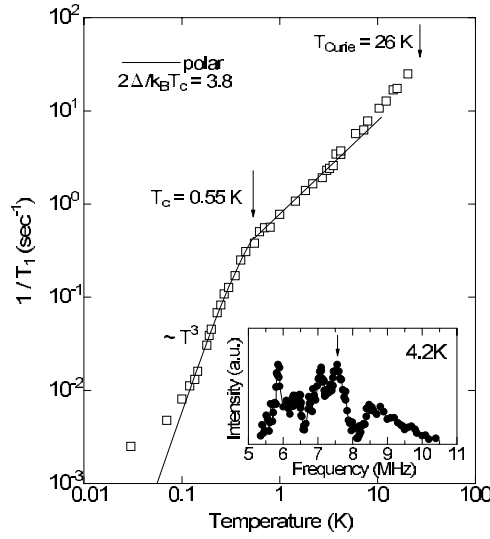
It is now quite controversial whether or not the SC and FM coexist at the microscopic level. It has been reported that the SC and FM are inhomogeneously separated and compete with each other [6]. Nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) are powerful tools enabling us to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

## 2. Experimental results

A polycrystalline sample with enriched  $^{73}\text{Ge}$  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  $^{73}\text{Ge}$  at  $P = 0$ , respectively. Since  $\text{UGe}_2$  has a strong uniaxial magnetic anisotropy, the sample is expected to be oriented along the easy axis, that is, the  $a$ -axis [7].

While  $\text{UGe}_2$  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  $T_1$ -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  $T_1T = \text{constant}$  below  $\sim 20$  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 UGe<sub>2</sub>. The solid curve was calculated by assuming a polar-type superconducting gap of  $2\Delta/k_B T_c$  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  $\sim 7.6$  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  $1/T_1 T = \text{constant}$  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 UGe<sub>2</sub> 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  $2\Delta/k_B T_c \sim 3.8$ , as indicated by the solid line. The observation of  $T_1 T = \text{constant}$ -like behaviour below 100 mK, however, prevents us from determining any precise gap structure. A future issue is hence whether the  $T_1 T = \text{constant}$ -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  $^{73}(1/T_1)$  probes both anomalies inherent in the FM and SC.

### 3. Summary

We have reported the  $T$ -dependence of  $1/T_1$  via  $^{73}\text{Ge}$  NMR/NQR measurements on the ferromagnetic superconductor UGe<sub>2</sub> under  $P = 1.3$  GPa.  $1/T_1 T = \text{constant}$  behaviour is observed well below  $T_{Curie}$ , whereas it is enhanced as the temperature approaches  $T_{Curie}$ . The bulk nature of the SC in UGe<sub>2</sub> 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  $T_1 T = \text{constant}$ -like behaviour below 100 mK does not allow us to propose any promising model for the SC in UGe<sub>2</sub>.

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  $UGe_2$ .

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