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Physica B 359-361 (2005) 1057-1059



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## Evidence for the microscopic coexistence of superconductivity and ferromagnetism in UGe<sub>2</sub>:<sup>73</sup>Ge-NMR/NQR study

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## Abstract

We report on the pressure-induced novel phases of ferromagnetism and superconductivity (SC) in the itinerant ferromagnet UGe<sub>2</sub> via the <sup>73</sup>Ge-NQR measurements under pressure (*P*). The NQR spectrum has revealed that the ferromagnetic phases are separated into weakly and strongly polarized phases around a critical value of  $P_c^* \sim 1.2$  GPa, pointing to a first-order transition around  $P_c^*$ . Here we present further evidence for the phase separation into ferromagnetic and paramagnetic phases around a critical pressure  $P_c \sim 1.6$  GPa. The measurements of nuclear spin–lattice relaxation rate  $1/T_1$  have probed that SC sets only in the ferromagnetic phase at  $T_{sc} \sim 0.2$  K, but it does not in the paramagnetic phase.

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Keywords: UGe2; NMR/NQR; Superconductivity; Ferromagnetism

Recently, superconductivity (SC) has been discovered in the background of ferromagnetism (FM) below the Curie temperature  $T_{\text{Curie}} = 52 \text{ K}$  in UGe<sub>2</sub> [1,2], ZrZn<sub>2</sub> [3], URhGe [4]. Since SC and FM are generally thought to be exclusive of each other, a great deal of surprise and interest are drawn. In UGe<sub>2</sub> with  $T_{\text{Curie}} = 52 \text{ K}$  at ambient pressure (P = 0), *P*-induced SC was discovered to emerge under P = 1 - 1.6 GPa, exhibiting a highest transition temperature  $T_{\text{sc}} \sim 0.8 \text{ K}$  at  $P_{\text{c}}^* \sim 1.2 \text{ GPa}$  [1,2]. It is noteworthy that the SC in UGe<sub>2</sub>

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<sup>0921-4526/\$ -</sup> see front matter © 2005 Published by Elsevier B.V. doi:10.1016/j.physb.2005.01.283

disappears above  $P_c \sim 1.6$  GPa, beyond which FM is suppressed. This fact implies that the SC and the FM in this compound may be cooperative phenomena. Indeed, the Ge-NQR measurements have shown the uniform coexistent phase of FM and unconventional SC with a line-node gap [5]. Here we report on microscopic characteristics of magnetism and SC at the pressure near  $P_c \sim$ 1.6 GPa in UGe<sub>2</sub> revealed by NQR measurements of enriched <sup>73</sup>Ge under *P*.

A polycrystalline sample enriched with <sup>73</sup>Ge was prepared and crushed into powder to allow a maximal penetration of oscillating magnetic field into the sample. Our NQR spectrum reveals a structure consisting of well-separated peaks associated with three inequivalent Ge sites in a unit cell. Fig. 1(a) indicates the spectrum for the PM phase at T = 35 K and P = 1.5 GPa close to  $P_c \sim$ 1.6 GPa where FM disappears. The spectrum in f = 8.2-8.8 MHz arises from Ge1 and Ge2. Here note that the two NQR lines for the Ge1 site are almost overlapped due to the asymmetry parameter  $\eta \sim 1$ . As temperature decreases below 10 K, the spectral shape is slightly broadened as seen for the spectrum at T = 4.2 K in Fig. 1(b), although the overall shape remains unchanged, suggesting that the system remains mostly in the PM state. This result reveals that the phase separation occurs into the PM and ferromagnetic phases. Here we focus on the change in NQR spectrum under the presence of  $H_{int}$  at the Ge1 site due to the onset of FM. This is because the Ge1 site is closely located to the uranium zig-zag chain responsible for FM and then the number of Gel sites is twice that of the Ge2 and Ge3 sites in a unit cell. In fact, the NQR spectrum that consists of two peaks for the Gel site at the PM state in Fig. 1(a) is split into multi-NQR lines in Fig. 1(b). From the comparison between experiment and calculation, a fraction of the PM to ferromagnetic phase is estimated to be 5:2 and the internal field at the Ge1 site as  $H_{\text{int}} = 0.2 \text{ T}$  at T = 4.2 K. Thus, the first-order



Fig. 1. The NQR spectrum in f = 8.1-9 MHz at P = 1.5 GPa and (a) T = 35 K for the PM phase, (b) the spectrum at T = 4.2 K is well simulated by assuming that the phase separation into the PM and ferromagnetic phases takes place with a fraction of 5:2 and the internal field  $H_{int} = 0.2$  T for the FM.



Fig. 2. The *T* dependence of  $1/T_1$  for the paramagnetic and ferromagnetic phases. The long component in  $1/T_1$  for the ferromagnetic phase indicates that the SC sets at  $T_{\rm sc} \sim 0.2$  K, but the short one does not for the PM phase.

transition from the ferromagnetic to PM phase was evidenced around  $P_c \sim 1.5$  GPa from the microscopic point of view, as for the case around  $P_c^* \sim 1.2$  GPa where the phase separation emerges into the weakly and strongly polarized ferromagnetic phases below  $T^*$  [6]. Fig. 2 shows the T dependence  $1/T_1$  at P = 1.5 GPa and H = 0. As temperature decreases below 10 K, two components in  $T_1$  appear associated with the PM and ferromagnetic phases, suggesting that FM sets in. This result is corroborated by the fact that the NQR spectrum probes the phase separation into the PM and ferromagnetic phases below  $T_{\text{Curie}} \sim$ 15 K. Note that a single component above 10 K is dominated by FM fluctuations. So  $T_{\text{Curie}} \sim$ 10–15 K may be estimated. It is noteworthy that the long component in  $T_1$  due to the ferromagnetic phase is shown to decrease below  $T \sim 0.2$  K which may be ascribed to a superconducting transition temperature for FM. This is because the presence of  $T_{\text{sc}}$  is actually confirmed in the phase diagram. Markedly, SC seems to be absent in the PM phase, assuring on a microscopic level that the SC in UGe<sub>2</sub> emerges under the background of FM.

In summary, the <sup>73</sup>Ge-NQR measurements on UGe<sub>2</sub> at P = 1.5 GPa have revealed that the phase separation into ferromagnetic and PM phases takes place near  $P_c \sim 1.6$  GPa, nevertheless the SC sets at  $T_{sc} \sim 0.2$  K only in the ferromagnetic phase, but not in the PM phase.

This work was supported by a Grant-in-Aid for Creative Scientific Research (15GS0123), MEXT and The 21st Century COE Program supported by the Japan Society for the Promotion of Science.

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