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### Permalink

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### Journal

Journal of the Korean Physical Society, 63(3)

### ISSN

0374-4884

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### Publication Date

2013-08-01

### DOI

10.3938/jkps.63.627

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# Magnetic Property in the Ferromagnetic Superconductor $\text{UGe}_2$ at Pressures above the Ferromagnetic Critical Pressure

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(Received 31 May 2012)

We have studied the high-pressure magnetic property in  $\text{UGe}_2$  where ferromagnetic superconductivity appears under high pressure. In this study, we focus on the magnetic property at pressures above the ferromagnetic critical pressure  $P_c = 1.6$  GPa. The temperature and magnetic field dependences of the dc-magnetization have been measured under high pressures up to 5.1 GPa by using a ceramic anvil high pressure cell. At pressures above  $P_c$ , the magnetic susceptibility  $\chi$  shows a broad maximum around  $T_{\chi_{max}}$  and the magnetization at 2.0 K shows an abrupt increase (metamagnetic transition) at  $H_c$ . With increasing pressure, the peak structure in  $\chi$  becomes broader, and the peak position  $T_{\chi_{max}}$  moves to the higher temperature region. The metamagnetic field  $H_c$  increases rapidly with increasing pressure. At pressures above 4.1 GPa,  $\chi$  shows a simple temperature dependence, and the magnetization increases linearly with increasing field. These phenomena in  $\text{UGe}_2$  resemble to those in the intermetallic compounds of  $3d$  transition metals such as  $\text{Co}(\text{S}_{1-x}\text{Se}_x)$  and  $\text{YCo}_2$ . We discuss the experimental results by using the phenomenological spin-fluctuation theory.

PACS numbers: 75.50.Cc, 74.20.Mn, 74.62.Fj

Keywords: Ferromagnetic superconductor, Metamagnetic transition, Spin fluctuations

DOI: 10.3938/jkps.63.627

## I. INTRODUCTION

The coexistence of ferromagnetism and superconductivity has been an important topic in condensed matter physics from both theoretical and experimental points of view [1]. Ferromagnetic superconductivity has been discovered in uranium compounds such as  $\text{UGe}_2$ ,  $\text{URhGe}$ , and  $\text{UCoGe}$  [2–5]. The ferromagnet  $\text{UGe}_2$  is the first compound where the superconductivity was discovered within its ferromagnetic phase. Figure 1 shows the pressure-temperature phase diagram in  $\text{UGe}_2$  from previous studies [6]. The Curie temperature  $T_C(P)$  decreases with increasing pressure from  $T_C = 53$  K at ambient pressure, the ferromagnetic transition changes from a second to first order at a tricritical point (TCP), and superconductivity appears at pressures above 1.0 GPa.

The bulk nature of the ferromagnetic superconductivity was confirmed by the specific heat measurements under high pressure [7,8]. The emergence of superconductivity is clearly related to an additional boundary,  $T_x(P)$ , that splits the ferromagnetic phase into a low-pressure FM2 and a high-pressure FM1 region. At  $T = 0$ , the transition from FM2 to FM1 is also first order and occurs at a pressure  $P_x \sim 1.2$  GPa where the ordered moment  $\mu_0$  drops from 1.4 to  $0.9 \mu_B/\text{U}$  and where the electronic specific-heat coefficient jumps from 50 to about  $100 \text{ mJ mol}^{-1} \cdot \text{K}^{-2}$  [8–12]. The first order transition at  $T_x(P)$  changes to a crossover through a critical endpoint at  $T_{CEP}$ . The transition from FM1 to paramagnetism (PM) at the critical pressure  $P_c = 1.6$  GPa is first order at zero temperature.

We have studied  $\text{UGe}_2$  by using various experimental methods [7–11]. In this study, we focus on the magnetic property at pressures above  $P_c$  where the ground state is paramagnetic. Previous studies up to 1.8 GPa showed

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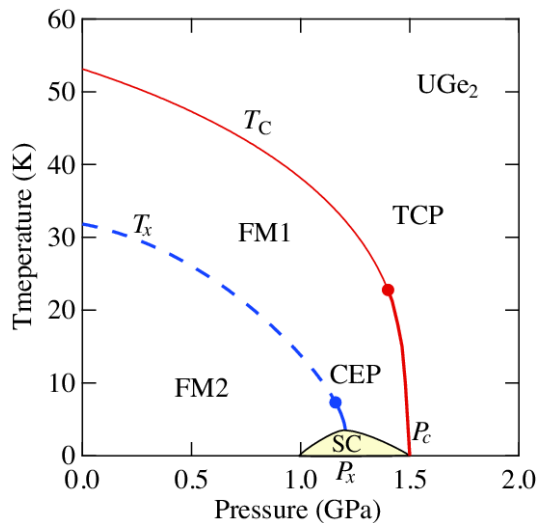


Fig. 1. (Color online) Temperature-pressure phase diagram in  $\text{UGe}_2$ . Thick lines represent first-order transitions and thin line denotes second-order transition. The dashed line indicates a crossover, and the dots mark the positions of critical points.

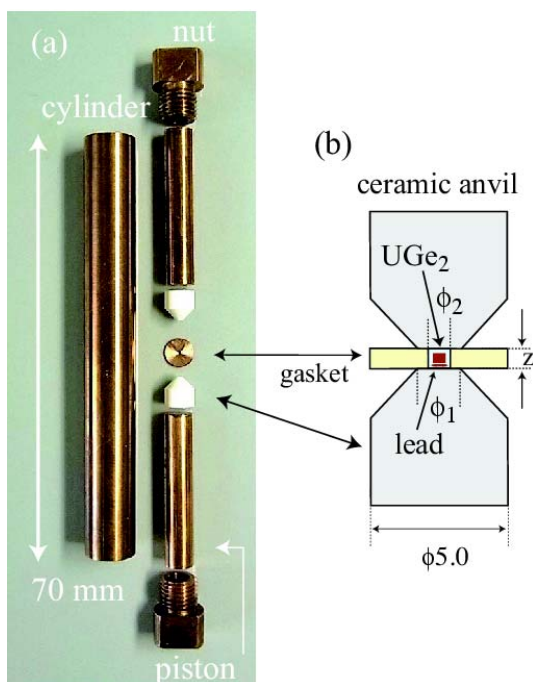


Fig. 2. (Color online) (a) Photograph of the miniature ceramic anvil high pressure cell (mCAC). (b) Schematic illustration of the Cu-Be gasket and ceramic anvils in the mCAC.

that a broad maximum appears in the temperature dependence of the ac magnetic susceptibility and that a ferromagnetic state is induced at the metamagnetic field  $H_c$  above  $P_c$  [11–13]. We measured the dc magnetization in  $\text{UGe}_2$  up to 5.1 GPa with newly developed high pressure cell.

## II. EXPERIMENTS AND DISCUSSION

We measured the magnetization  $\text{UGe}_2$  under high pressure with a miniature ceramic anvil cell (mCAC) [14]. Figure 2 shows (a) a photograph of the mCAC and (b) a schematic illustration of the Cu-Be gasket and ceramic anvils in the cell. The cell was developed for magnetic measurements with a commercial superconducting quantum interference (SQUID) magnetometer MPMS (Magnetic Property Measurement System) from Quantum Design (USA) [15]. The mCAC is 8.7 mm in diameter and 70 mm in length. The long cylinder is designed to reduce the contribution from the cylinder and the locknuts constructed out of a nonmagnetic hardened Cu-Be alloy. A Cu-Be gasket separates two opposing anvils made of the composite ceramic FCY20A produced by the Fuji Die company, Japan [14]. The composite ceramic is a mixture of  $\text{Y}_2\text{O}_3$ -partially stabilized zirconia ( $\text{ZrO}_2$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) synthesized under high pressure and temperature. We used a high-quality single-crystal sample of  $\text{UGe}_2$  grown by the Czochralski pulling method in a tetra-arc furnace [11]. The culet size of the ceramic anvil for the present study was  $\phi_1 = 1.0$  mm. The sample and a lead (Pb) pressure manometer were loaded in a sample chamber whose diameter was  $\phi_2 = 0.50$  mm. The chamber was filled with the pressure-transmitting medium glycerin [16]. The thickness of the Cu-Be gasket was  $z = 0.50$  mm. The size of the single-crystal sample was  $0.30 \times 0.30 \times 0.40$  mm<sup>3</sup>. The magnetic field  $H$  was applied parallel to the magnetic easy axis (the [100] direction) of the orthorhombic crystal structure. The load was applied to the pressure cell through a piston and clamped by using a locking nut at room temperature. The cell was installed in the MPMS magnetometer and cooled to low temperatures. The SQUID response of the pressure cell was collected with and without the sample to obtain the magnetization of the sample. The difference signal was fitted to a calculated form assuming a point dipole moment and using a specially written external program to obtain the magnetization of the sample. The pressure values at low temperatures were determined from the pressure dependence of the superconducting transition temperature in lead [17–19].

To confirm the performance of the mCAC, we measured the magnetization,  $M(T)$ , in the ferromagnetic phases FM1 and FM2 in  $\text{UGe}_2$ . Figures 3(a) and (b) show the temperature dependences of the magnetization at 1 bar, 0.75 GPa and 1.40 GPa under magnetic fields of 1.0 and 0.01 T along the  $a$ -axis (easy axis), respectively. With decreasing temperature, the magnetization  $M(T)$  increases largely at temperatures below the Curie temperature  $T_C$ . The magnetization shows an additional anomaly around the crossover temperature  $T_x$  from the FM1 to the FM2. The values of  $T_C$  and  $T_x$  were determined from the peak position in the temperature dependence of  $-dM/dT$  shown by arrows in Fig. 3. The pres-

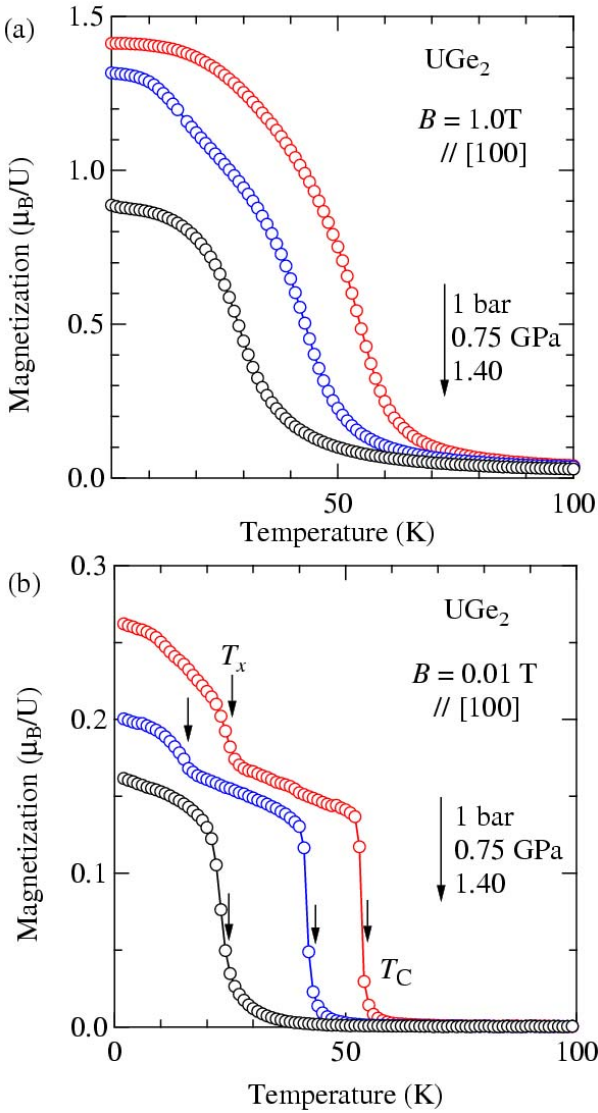


Fig. 3. (Color online) Temperature dependence of the magnetization  $M(T)$  at 1 bar, 0.75 and 1.40 GPa under magnetic fields of (a) 1.0 and (b) 0.01 T along the  $a$ -axis (easy axis) in UGe<sub>2</sub>.

sure dependences of  $T_C$  and  $T_x$ , the value of the magnetic moments in the FM1 and 2 phases are consistent with those in previous studies [9,12].

Next, we measured the temperature dependence of the magnetic susceptibility  $\chi(T)$  under magnetic field of 1.0 T at pressures above the critical pressure  $P_c$  as shown in Fig. 4. The susceptibility shows a broad maximum around  $T_{\chi max} = 27.5$  K, which is qualitatively consistent with a previous study by the ac magnetic susceptibility measurement up to 1.76 GPa [13]. With increasing pressure, the peak structure becomes weaker and  $T_{\chi max}$  increases to 34.7 K at 2.70 GPa. The low-temperature magnetic susceptibility decreases with increasing pressure. Above 3.9 GPa, the peak structure in  $\chi(T)$  disappears, and the susceptibility shows a monotonic temper-

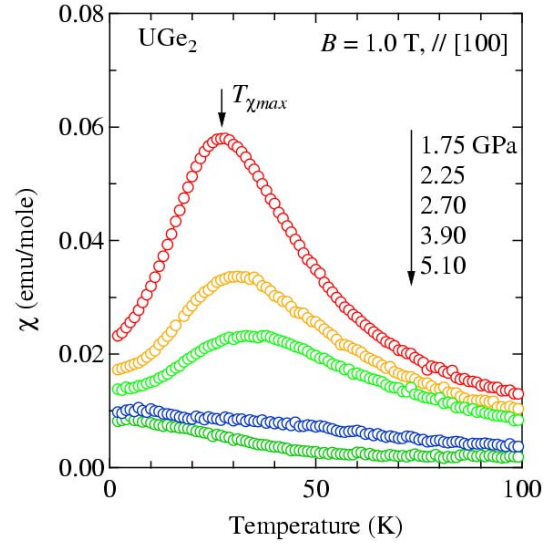


Fig. 4. (Color online) Temperature dependence of the magnetic susceptibility  $\chi(T)$  under a magnetic field of 1.0 T at pressures above the ferromagnetic critical pressure  $P_c$  in UGe<sub>2</sub>.

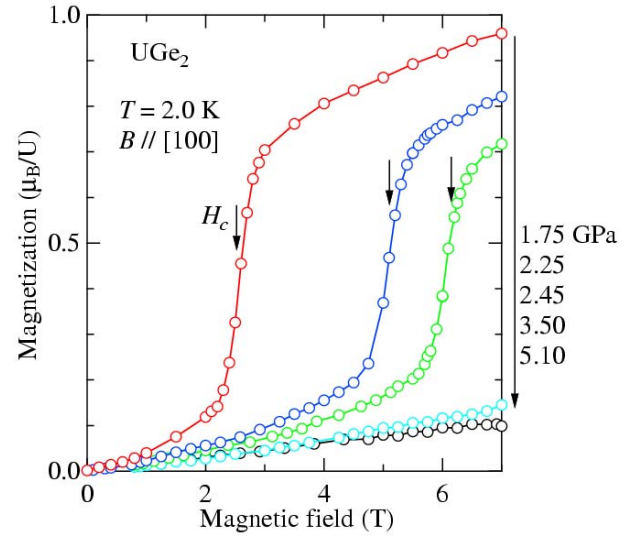


Fig. 5. (Color online) Magnetic field dependence of the magnetization  $M(H)$  at 2.0 K at pressures above the ferromagnetic critical pressure  $P_c$  in UGe<sub>2</sub>.

ature dependence.

Figure 5 shows the field dependence of the magnetization measured at 2.0 K above  $P_c$ . At 1.75 GPa, the magnetization shows a discontinuous increase (metamagnetic transition) at  $H_c = 2.4$  T. The field-induced transition from the paramagnetic to the ferromagnetic states in UGe<sub>2</sub> is consistent with a previous study up to 2.0 GPa [11,12]. With increasing pressure, the metamagnetic transition field is increased to 6.0 T at 2.45 GPa. Above that pressure,  $H_c$  may be higher than the maximum magnetic field of 7 T in the MPMS. The magnetization increases linearly with increasing magnetic field

at 3.50 and 5.10 GPa. The monotonic field dependence of the magnetization seems to be related to the disappearance of the broad maximum in the magnetic susceptibility.

Itinerant electron metamagnetism or a broad maximum in the magnetic susceptibility has been observed in intermetallic compounds of  $3d$  transition metals such as  $\text{Co}(\text{S}_{1-x}\text{Se}_x)$  and  $R\text{Co}_2$  with  $R = \text{Y}$  and  $\text{Lu}$  [20,21]. Theretically, the metamagnetic transition can occur in a self-consistent spin fluctuation theory when  $b < 0$  in a Ginzburg-Landau expansion of the free energy to sixth order in  $M$ :  $\Delta F = \frac{1}{2}aM^2 + \frac{1}{4}bM^4 + \frac{1}{6}cM^6$  [22]. The equation of state for  $M$  and the magnetic field  $B$  is given by  $B = aM + bM^3 + cM^5$ , where  $a$  is the inverse magnetic susceptibility at  $T = 0$  K;  $a = \chi(0)^{-1}$ . The coefficients  $a$ ,  $b$  and  $c$  are functions of the electronic density of states and its derivatives at the Fermi energy.  $b < 0$  indicates negative mode-mode coupling among spin fluctuations. The theory predicts that  $T_C$  decreases as the parameter  $ac/b^2$  is increased and that the transition changes from second to first order at a particular value of  $ac/b^2 = 5/28$ . Above that value,  $T_C$  decreases quickly, and the ground state becomes paramagnetic above  $ac/b^2 = 3/16$ . When  $a > 0$ ,  $b < 0$ ,  $c > 0$  and  $3/16 < ac/b^2 < 9/20$ , the paramagnetic state is stable at zero magnetic field, but the ferromagnetic state becomes stable at high magnetic fields. The field-induced metamagnetic transition from the paramagnetic to the ferromagnetic state occurs at a critical field  $H_c$ . If we assume that  $ac/b^2$  is an increasing function of the pressure, the pressure change of the ground state in  $\text{UGe}_2$  and the appearance of the metamagnetic transition above  $P_c$  can be understood from this theoretical point of view.

At a finite temperature  $T$ , the coefficients  $a$ ,  $b$  and  $c$  are renormalized by thermal spin fluctuations, and the equations of state is written as  $B = A(T)M + B(T)M^3 + C(T)M^5$  [22]. The coefficients  $A(T)$ ,  $B(T)$ , and  $C(T)$  are functions of  $a$ ,  $b$ ,  $c$  and the mean-square local amplitude of thermal fluctuation moment  $\xi(T)$ . The inverse magnetic susceptibility is given by the relation:  $\chi^{-1}(T) = a + \frac{5}{3}b\xi(T)^2 + \frac{35}{9}c\xi(T)^4$ . When  $a > 0$ ,  $b < 0$ ,  $c > 0$  and  $5/28 < ac/b^2 < 9/20$  which corresponds to the pressure region above the tricritical pressure  $P_{TCP}$  in  $\text{UGe}_2$ , the magnetic susceptibility increases and then decreases through a maximum value with increasing temperature. With increasing temperature, the amplitude of the longitudinal spin fluctuation is enhanced due to the negative mode-mode coupling among the spin fluctuations, causing the maximum in the magnetic susceptibility  $\chi$ . The spin-fluctuation theory for the itinerant electron metamagnetism can explain the present results in  $\text{UGe}_2$ . For a more quantitative analysis, detailed measurements and analyses are necessary. These are left for a future study.

## ACKNOWLEDGMENTS

This work was supported by a Grant-in-Aid for Scientific Research on Innovative Areas "Heavy Electrons" (Nos. 20102002 and 23102726), Scientific Research S (No. 20224015), A(No. 23246174), and C (No. 21540373 and 22540378), and for Young Scientists (B) (No. 2274021) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) and the Japan Society of the Promotion of Science (JSPS).

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