Tricritical point of a ferromagnetic transition in UGe$_2$

N Kabeya$^1$, R Iijima$^1$, E Osaki$^1$, S Ban$^1$, K Imura$^1$, K Deguchi$^1$, N Aso$^2$, Y Homma$^3$, Y Shiokawa$^3$ and N K Sato$^1$

1Department of Physics, Graduate School of Science, Nagoya University, Nagoya 464-8602, Japan
2Department of Physics and Earth Science, Faculty of Science, University of the Ryukyus, Okinawa 903-0213, Japan
3Institute for Materials Research, Tohoku University, Ibaraki 980-0213, Japan

E-mail: *kabeya.noriyuki@d.mbox.nagoya-u.ac.jp*

Abstract. Thermal expansion and magnetostriction measurements of the superconducting ferromagnet UGe$_2$ under pressure were carried out. The temperature dependence of the thermal expansion coefficient shows a peak at the Curie temperature. When pressure is varied, the peak exhibits a maximum in the vicinity of a tricritical point (TCP), which separates the second-order phase transition from the first-order transition. From results of these measurements, we first construct the magnetic phase diagram including the TCP ($P_{TCP}$: 5 kbar). We also show that two lines characterizing the metamagnetism and the magnetic susceptibility emerge from the TCP. We argue that these magnetic properties in the vicinity of the TCP can be understood within a phenomenological frame of spin fluctuations.

1. Introduction

The ferromagnet UGe$_2$ has attracted much attention since the discovery of superconductivity under pressure [1, 2]. As the superconductivity vanishes at the same critical pressure $P_C$ ($\sim 15$ kbar) as the ferromagnetism, it is believed that there is a strong coupling between them. In the present study, we focus on magnetic properties of this intriguing material.

UGe$_2$ crystallizes in the orthorhombic ZrGa$_2$-type structure (space group Cmmm) with lattice constants $a = 4.036$ Å, $b = 14.928$ Å, and $c = 4.116$ Å [3]. At ambient pressure, the ferromagnetic phase transition occurs at $T_C \sim 53$ K. With increasing pressure, the Curie temperature $T_C$ monotonically decreases and finally collapses to zero at $P_C$. It was suggested that the ferromagnetic transition changes from second order to first order at a certain pressure slightly below $P_C$ [4, 5]. This means that there is a tricritical point (TCP) on the $T_C(P)$ curve of the $P$-$T$ phase diagram. It was also reported that a metamagnetic transition appears in the paramagnetic phase. Furthermore, a maximum emerges in the temperature dependence of the magnetic susceptibility [4, 6]. These characteristics are often observed in itinerant ferromagnets, and can be readily understood within a phenomenological framework [7, 8]. Let us write down the magnetic equation of state for the bulk moment $M$ and the external magnetic field $H$ as follows [7, 8],

$$H = A(T)M + B(T)M^3 + C(T)M^5,$$

where

$$A(T) = a + \frac{5}{3}b\xi(T)^2 + \frac{35}{9}c\xi(T)^4, \quad B(T) = b + \frac{14}{3}c\xi(T)^2, \quad C(T) = c.$$
Here \( a, b, \) and \( c \) denote the Ginzburg-Landau expansion coefficients of the free energy. \( \xi(T)^2 \) is the mean square amplitude of spin fluctuations and considered to be a monotonically increasing function of temperature. A simple calculation yields \( \xi(T_C), \xi(T_{\text{max}}), \) and \( \xi(T_0) \) as a function of \( ac/b^2 \), where \( T_{\text{max}} \) and \( T_0 \) indicate a temperature at which the magnetic susceptibility shows maximum and the metamagnetic transition disappears, respectively. If the quantity \( \xi(T)^2 \) is known as a function of temperature, these characteristic temperatures are uniquely determined.

![Figure 1](image)

Figure 1. Schematic phase diagram taken from ref.\[7\]. \( a, b, \) and \( c \) denote the G-L coefficients of the free energy expansion. The mean square amplitude of spin fluctuations \( \xi(T)^2 \) is expected to be a monotonically increasing function of temperature. \( T_C, T_{\text{max}}, \) and \( T_0 \) indicate the Curie temperature, a temperature at which the magnetic susceptibility shows maximum, and a temperature at which the metamagnetic transition disappears, respectively.

In Figure 1, we schematically illustrate \( \xi(T)^2 \) as a function of \( ac/b^2 \), which represents the magnetic phase diagram. Here, the parameter \( ac/b^2 \) may be regarded as a function of pressure. Note that \( T_C < T_0 < T_{\text{max}} \) in the first order transition region. Interestingly, it is expected from the phase diagram that \( T_{\text{max}} \) line, a temperature at which the magnetic susceptibility shows maximum, emerges from the TCP. For UGe\(_2\), a number of \( P-T \) phase diagrams were reported to date \[2, 9, 10, 11, 12\], but a complete phase diagram including a tricritical point is unknown yet. Here we present the magnetic phase diagram near the TCP.

2. Experimental results and discussion

A single crystalline sample of UGe\(_2\) was grown by the Czochralski pulling method in a tetra-arc furnace. Hydrostatic pressure was applied using a NiCrAl-BeCu piston cylinder cell with Dalpne oil 7373 as a pressure transmitting medium. In order to measure the thermal expansion under pressure, we employed the active-dummy technique using strain gages (SKF-5414, KYOWA) with a copper block (6N purity) as a reference material. The strain gages were glued on clean surfaces of the UGe\(_2\) sample so as to measure the dilatation along the orthorhombic \( a, b, \) and \( c \)-axes. In this paper, we present only data of the dilatation along the \( b \)-axis that showed the largest anomaly at the Curie temperature; other data along the \( a \) and \( c \)-axes will be given elsewhere. All these experimental settings are the same as those in our previous experiment \[13\].

Figure 2 shows the temperature dependence of the thermal expansion coefficient \( \alpha(T) \) at pressures up to 14.8 kbar, that is close to but still less than \( P_C \). The negative peak is ascribed to the ferromagnetic transition. At \( P = 0.5 \) kbar, the transition is mean-field like reflecting the second-order transition. As pressure increases, \( T_C \) decreases monotonically, and the peak becomes sharper. Note that the peak height shows a maximum as a function of pressure at approximately \( P \sim 13 \) kbar (see the inset). This characteristic pressure is identified as a tricritical pressure \( P_{TCP} \). With further increasing pressure, the peak height turns to decrease, and the peak shape becomes symmetric with respect to temperature, which we ascribe to the first-order transition broadened by some inhomogeneity.

Figure 3 shows the magnetic field dependence of the dilatation (the forced magnetostriction) at 14.5 kbar (< \( P_C \)) and several temperatures. The magnetic field was applied along the \( a \)-axis, the magnetization easy axis. At \( T = 33 \) K, we observe the dilatation to monotonically increase with \( H \). At low temperatures below about 27 K, it shows a rather steep increase at
a certain field, for instance approximately 0.4 T at 23 K. This increase, which we ascribe to the metamagnetization, becomes prominent with lowering temperature. At temperatures below about 17 K, the anomaly is observed at zero field, meaning that the first order ferromagnetic transition occurs there.

Figure 4 represents the resulting magnetic phase diagram deduced from the thermal expansion and the magnetostriction data. $T_0$ was obtained from the magnetostriction measurement; the magnetostriction coefficient $\lambda (=1/L(\partial L/\partial H))$ exhibits a peak as a function of $H$, and its width increases above $T_0$, for instance, $T_0 \sim 24$ K at $P = 14.5$ kbar (see Figure 3). The temperature $T_0$ gradually decreases with increasing pressure. We also plot $T_{\text{max}}$ deduced from a temperature dependence of the magnetic susceptibility (not shown here). The TCP is identified as an inflection-point like anomaly of the $T_C(P)$ curve, i.e., $P_{\text{TCP}} \sim 12.5$ kbar. It is noteworthy that all the lines ($T_C$, $T_{\text{max}}$, $T_0$) appear to merge at the TCP, as is expected from the phenomenological model of the spin fluctuations.

3. Summary
We measured the thermal expansion and magnetostriction of the superconducting ferromagnet UGe$_2$ under pressure. The anomaly of the thermal expansion coefficient provides the Curie temperature as a function of pressure. Remarkably, it exhibits maximum in the vicinity of the tricritical point (TCP), $P_{\text{TCP}} \sim 12.5$ kbar, at which the second order phase transition
Figure 4. Magnetic phase diagram in the vicinity of the tricritical point (TCP); TCP is a characteristic point in the \( P-T \) coordinate that separates the second-order phase transition from the first-order one. The Curie temperature \( T_C \) was defined as a temperature at which the thermal expansion coefficient shows a peak. \( T_{\text{max}} \) indicates a temperature at which the temperature dependence of the magnetic susceptibility exhibits maximum. \( T_0 \) denotes a temperature above which the metamagnetism disappears.

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