NATURE OF FERROMAGNETIC PHASE IN PRESSURE-INDUCED FERROMAGNETIC SUPERCONDUCTOR UGe₂*

T. NISHIOKA, G. MOTOYAMA, S. NAKAMURA AND N.K. SATO

Department of Physics, Nagoya University, Chikusa-ku, Nagoya 464-8602, Japan

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We have measured isothermal magnetization curves of the ferromagnetic superconductor UGe₂ at pressures up to 18.3 kbar and at temperatures down to 0.45 K. The pressure dependence of the Curie temperature T_{Curie} evaluated from a linear part of the Arrott plot is relatively in agreement with the literatures. The paramagnetic Curie temperature, however, deviates from T_{Curie} with increasing pressure, especially above about 10 kbar. In addition, we have found a staircase-like magnetization hysteresis loop with regular interval of magnetic fields only below about 1 K. We ascribe it to the macroscopic quantum tunneling, and the analysis suggests tiny magnetic domain formation whose size is smaller than the superconducting coherence length.

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1. Introduction

The ferromagnet UGe₂ exhibits superconductivity in the pressure range between ~10 and ~16 kbar [1]. Since the ferromagnetism disappears nearly at the same pressure ~16 kbar, the superconducting phase exists completely in the ferromagnetic phase. It seems to be assumed that both orderings arise from 5f electrons in U atom, and that the ferromagnetism and the superconductivity coexist homogeneously. However, our recent ac magnetic susceptibility measurements clarified that the superconductivity is inhomogeneous in the real space [2]. We have also implied that the ferromagnetism becomes inhomogeneous at high pressures where the superconducting phase appears.

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The purpose of the present paper is to investigate detailed nature of the ferromagnetic phase, especially at pressures above 10 kbar and at low temperatures below 1 K.

Single crystals were grown by the Czochralski pulling method using a tetra-arc furnace. The magnetization was measured by applying the field along the *a*-axis using a laboratory-made vibrating sample magnetometer, and the sample was immersed in liquid ³He. The pressure was generated by a beryllium-copper piston-cylinder clamp device using Fluorinert as a pressure transmitting medium.

2. Results and discussion

Fig. 1 shows the field dependence of the magnetization at 4.5 K for several selected pressures. At ambient pressure the magnetization saturates at low fields, while it hardly saturates at high pressures. An Arrott plot $(M^2 vs H/M \text{ plot})$ gives T_{Curie} at which an extrapolated straight line passes through the origin. Below T_{Curie} the longitudinal intercept of the Arrott plot gives the square of spontaneous magnetization M_0 , and above T_{Curie} the transverse intercept gives the inverse of magnetic susceptibility χ . The low pressure data in the Arrott plot lie on a straight line, while the high pressure data especially above 15 kbar tend to deviate from a straight line of the plot at low fields.



Fig. 1. Magnetization as a function of magnetic field at 4.5 K for several selected pressures.

Fig. 2 shows M_0 and $1/\chi$ as a function of temperature at several pressures estimated from the Arrot plot except the very low field data. The Curie temperature obtained in this way is in good agreement with the peak temperature of the ac susceptibility that was measured at the same time (not shown). The pressure dependence of T_{Curie} relatively agrees with that in the literature [3–5]. At ambient pressure, the paramagnetic Curie temperature T_{f} is close to T_{Curie} as can be seen from Fig. 2, while T_{f} deviates from T_{Curie} with increasing pressure. This suggests that ferromagnetic fluctuation above T_{Curie} evolves with pressure. The inverse susceptibility above 50 K for each pressure lies on a straight line, with nearly the same slope, corresponding to the effective moment $\mu_{\text{eff}} \sim 2.5 \mu_{\text{B}}$. In addition, at 16.9 kbar we observed the separation of the zero-field-cooled and field-cooled magnetization, which is similar to the spin glass behavior. These results suggest that the ferromagnetism is inhomogeneous at pressures above about 10 kbar.



Fig. 2. Temperature dependence of the spontaneous magnetization M_0 and inverse susceptibility $1/\chi$ which were obtained from the Arrot plot. The symbols are as follows: P = 0 kbar (•), 9.1 kbar (o), 12.3 kbar (\triangle), 12.9 kbar (\bigtriangledown), 15.0 kbar (\square) and 16.9 kbar (\diamondsuit).

Fig. 3 shows ferromagnetic hysteresis loops at several temperatures down to 0.45 K at 11.5 kbar just above which superconductivity appears. This measurement was done for a single crystal different from that used for measurements given in Figs. 1, 2. We see a smooth hysteresis curve at 4.4 K. As temperature is decreased down to 0.59 K, the shape of the hysteresis curve is unchanged, although the coercive force shows an increase. When the temperature is lowered to 0.45 K, the hysteresis loop changes quite suddenly into a staircase-like loop. The jump occurs regularly every ~ 0.03 T, as can be seen from the Fig. 3. Since these jumps were observed only at

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low temperatures below 0.5 K and with regular field interval, we ascribe these jumps to the macroscopic quantum tunneling (MQT) [7]. Although this type of magnetization jump was observed in a simple ensemble of noninteracting molecules such as $Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4$ [6], UGe₂ may be the first example showing MQT in macroscopic materials. An analysis based on MQT gives the domain size of about 40 Å(the detailed analysis is given in Ref. [7]), which is smaller than the superconducting coherence length 130–200 Å [5]. If the tiny domains are adversely oriented, then internal molecular fields due to the ferromagnetism can be canceled out in the scale of the coherence length.



Fig. 3. M vs H hysteresis loops of UGe₂ at low temperatures and at pressure of 11.5 kbar.

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