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

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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 $T_{\text{Curie}}$ evaluated from a linear part of the Arrott plot is relatively in agreement with the literatures. The paramagnetic Curie temperature, however, deviates from $T_{\text{Curie}}$ 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 $\sim 10$ and $\sim 16$ kbar [1]. Since the ferromagnetism disappears nearly at the same pressure $\sim 16$ 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 \( \alpha \)-axis using a laboratory-made vibrating sample magnetometer, and the sample was immersed in liquid \(^3\)He. 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 \( H/M \) plot) gives \( T_{\text{Curie}} \) at which an extrapolated straight line passes through the origin. Below \( T_{\text{Curie}} \) the longitudinal intercept of the Arrott plot gives the square of spontaneous magnetization \( M_0 \), and above \( T_{\text{Curie}} \) the transverse intercept gives the inverse of magnetic susceptibility \( \chi \). 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 Arrott 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 $T_{\text{Curie}}$ relatively agrees with that
in the literature [3–5]. At ambient pressure, the paramagnetic Curie tem-
perature $T_f$ is close to $T_{\text{Curie}}$ as can be seen from Fig. 2, while $T_f$ devi-
ates from $T_{\text{Curie}}$ with increasing pressure. This suggests that ferromagnetic fluc-
tuation above $T_{\text{Curie}}$ evolves with pressure. The inverse susceptibility above
50K for each pressure lies on a straight line, with nearly the same slope, cor-
responding to the effective moment $\mu_{\text{eff}} \sim 2.5\mu_B$. In addition, at 16.9kbar
we observed the separation of the zero-field-cooled and field-cooled magnet-
tization, 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 Arrott plot. The symbols are as
follows: $P = 0$ kbar (●), 9.1 kbar (○), 12.3 kbar (△), 12.9 kbar (▽), 15.0 kbar (□)
and 16.9 kbar (◇).

Fig. 3 shows ferromagnetic hysteresis loops at several temperatures down
to 0.45K at 11.5 kbar just above which superconductivity appears. This
measurement was done for a single crystal different from that used for mea-
surements given in Figs. 1, 2. We see a smooth hysteresis curve at 4.4K.
As temperature is decreased down to 0.59K, the shape of the hysteresis
curve is unchanged, although the coercive force shows an increase. When
the temperature is lowered to 0.45K, the hysteresis loop changes quite sud-
denly into a staircase-like loop. The jump occurs regularly every $\sim 0.03$ T,
as can be seen from the Fig. 3. Since these jumps were observed only at
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 non-interacting molecules such as Mn$_{12}$O$_{12}$(CH$_3$COO)$_{16}$(H$_2$O)$_4$ [6], UGe$_2$ 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 UGe$_2$ at low temperatures and at pressure of 11.5 kbar.

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