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Possible competitive coexistence of ferromagnetism and superconductivity in UGe_2

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We have measured magnetizations of the ferromagnet UGe_2 which shows superconductivity in a narrow external pressure range, $P_1 \leqslant P \leqslant P_2$, in the ferromagnetic state. When P is close to P_1 in the superconducting phase, the ac magnetic susceptibility indicates a peak-anomaly associated with the ferromagnetic transition and an imperfect superconducting shielding effect. As P increases away from P_1, the peak anomaly becomes substantially broad and obscure, while the diamagnetic susceptibility approaches a perfect superconducting shielding. We have also observed that a saturation magnetization at low temperature shows a steep decrease with increasing P in the superconducting state. From these observations, we suggest that the superconductivity coexists in a competitive way with the ferromagnetism; as P increases, a volume fraction of the superconducting state grows over the system, while the ferromagnetic ordering possibly becomes spatially inhomogeneous.

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Recently Saxena *et al.* discovered superconductivity in UGe_2 which emerges under high pressures between about 10 and 16 kbar.[1] UGe_2 is a ferromagnet with the Curie temperature T_{Curie} of about 53 K at ambient pressure. When an external pressure P is applied, T_{Curie} shows monotonic decrease with increasing pressure and seems to vanish at around 16 kbar, therefore the superconductivity appears only in the ferromagnetic phase.

In the 1970s coexistence of superconductivity (SC) and ferromagnetism (FM) was studied in rare earth compounds such as rare-earth rhodium borides.[2] In the case of $ErRh_4B_4$, for example, the system sets in superconducting phase at $T_{\mathrm{SC}} \sim 8.7$ K, and localized magnetic moments start to align ferromagnetically at $T_{\mathrm{Curie}} \sim 1.2$ K. The superconductivity finally disappears below about 0.9 K, thus there is a narrow temperature range in which the SC coexists with the FM. Thorough investigation for this compound revealed that the FM is carried by $4f$ electrons of Er atoms and the SC by $4d$ electrons of Rh atoms, which implies that SC and FM are separated in the real space. In the present case of UGe_2, however, both of the long-ranged ordered states are argued to be carried by $5f$ electrons of uranium atoms,[1,3] although it seems to us that there is no direct indication that the $5f$ electrons carry superconducting currents.

Other systems are known in which it is commonly accepted that both antiferromagnetism and superconductivity are carried by $5f$ electrons; UPd_2Al_3 is a typical example, in which magnetic excitons originating from a localized component of $5f$ electrons mediate superconducting pairing interaction between quasiparticles coming from an itinerant part of $5f$ electrons.[4] In UPd_2Al_3, magnetic moments are ferromagnetically aligned on a hexagonal basal-plane, which is stacked antiferromagnetically along a c-axis with a propagation wave vector $\mathbf{Q_0} = (0,0,1/2)$. Since a period of the alternative stacking of $2c \sim 8$ Å is much smaller than a superconducting coherence length of the order of 100 Å, internal fields (due to the antiferromagnetic ordering) which superconducting Cooper paired electrons may observe, are probably cancelled out. However, in ferromagnetism we expect that superconducting electrons detect a non-vanishing internal field. Thus, it is quite surprising that the FM with a local moment of the order of $1\mu_{\mathrm{B}}/\mathrm{U}$ coexists with the SC in UGe_2.

This interesting feature of UGe_2 raises a question concerning to the nature of the coexistence of FM and SC; are both of them homogeneous in the real space? To resolve this question we have made magnetization measurements in terms of ac and dc methods under external pressures.

A polycrystalline material was first prepared by melting stoichiometric amounts of natural uranium and germanium of high purity. Then single crystals were grown by the Czochralski pulling method using a tetra-arc furnace in a pure argon atmosphere. We did not make additional heat treatment. We characterized the grown single crystals using the X-ray, ac magnetic susceptibility, and electrical resistivity measurements. As some U-Ge binary compounds show ferromagnetism,[5,6] the ac magnetic susceptibility measurement is a good tool for detecting such ferromagnetic impurity phases. Combined results of the X-ray and ac magnetic susceptibility measurements showed no trace of secondary phases. The electrical resistivity measurements of the ingots indicated that a residual resistivity ratio is about 290 and 270 for samples named No. 2 and No. 3, respectively. (A current in the measurements flowed along a grown axis of the ingot.)

The dc magnetization measurements were carried out using a laboratory-made vibrating sample magnetometer (VSM) with a frequency of 0.5 Hz. The ac magnetic susceptibility was measured in terms of a conventional Hartshorn bridge circuit in a frequency range between 4 and 100 Hz. A peak-to-peak amplitude of a modulation field was ranged from ~ 0.06 to ~ 12 Oe. The sample was immersed in liquid ^{3}He and cooled down to about 0.35 K. The pressure was generated by means of a copper-beryllium (CuBe) clamp-type cylinder. We used a piston made of CuBe and tungsten carbide (WC) for the measurements of No. 2 and No. 3, respectively. In the case that the WC piston was used, we found a broad anomaly in the ac magnetic susceptibility at around 42 K, which was unambiguously ascribed to a contribution from the piston. Pressure transmitting medium was a 1:1 mixture of Fluorinert FC70 and FC77. We usually measure a superconducting transition temperature of indium to

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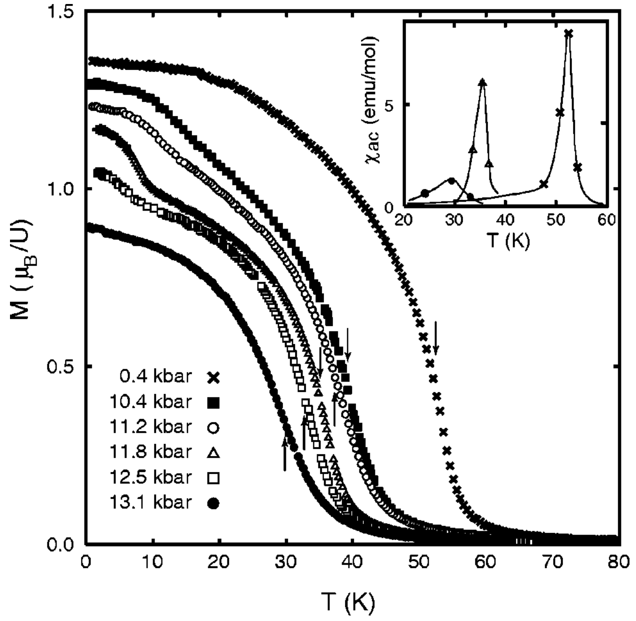

FIG. 1. Temperature dependence of dc magnetization $M(T)$ under external pressures for the sample No. 3. The inset shows the temperature dependence of the real part of ac magnetic susceptibility, $\chi_{\mathrm{ac}}(T)$, under external pressures. Note that a peak anomaly in $\chi_{\mathrm{ac}}(T)$ at a Curie temperature T_{Curie} is substantially damped at a pressure as large as 13.1 kbar. $M(T)$ displays an inflection at T_{Curie} (marked by an arrow) and a tail above it, because measurements were done by applying an external field of 2 kOe (parallel to the *a* axis). We also note that $M(T)$ shows an upturn behavior at around a characteristic temperature in an intermediate pressure range between about 10.4 and 12.5 kbar (see text in detail).

determine a pressure at low temperatures, but it did not work well in the present case, because a small magnetic field originating from the ferromagnetic sample affected the transition temperature of indium which was set close to the sample inside a cell made of Teflon. Therefore, we estimated the pressure by comparing a measured T_{Curie} of the sample with a curve of T_{Curie} vs P reported by Huxley *et al.*[3]

Figure 1 shows the temperature dependence of dc magnetization, $M(T)$, of No. 3 under external pressures. We also give in the inset the temperature dependence of the real part of ac magnetic susceptibility, $\chi_{\mathrm{ac}}(T)$, of the same sample at several pressures. $\chi_{\mathrm{ac}}(T)$ indicates a peak at the ferromagnetic Curie temperature T_{Curie}, as is usual and consistent with the results in the literature.[1] It should be noted that the peak is substantially damped at a pressure as large as 13.1 kbar. We found $M(T)$ to show a tail above T_{Curie} which is marked by an arrow. This is because the measurements of dc magnetization were made by applying a finite external magnetic field.

At ambient pressure, no apparent anomaly is visible in the $M(T)$ curve, but the application of the external pressure induces an upturn at around a characteristic temperature T_{M}. When we define T_{M} as a temperature at which the $M(T)$ curve shows an inflection, we obtain T_{M} as $\sim$14 K at 10.4 kbar. When P increases to 11.8 kbar, the upturn behavior becomes prominent with a lower T_{M} of $\sim$7.5 K. Further application of the pressure makes T_{M} to decrease and finally vanish at around 13 kbar.

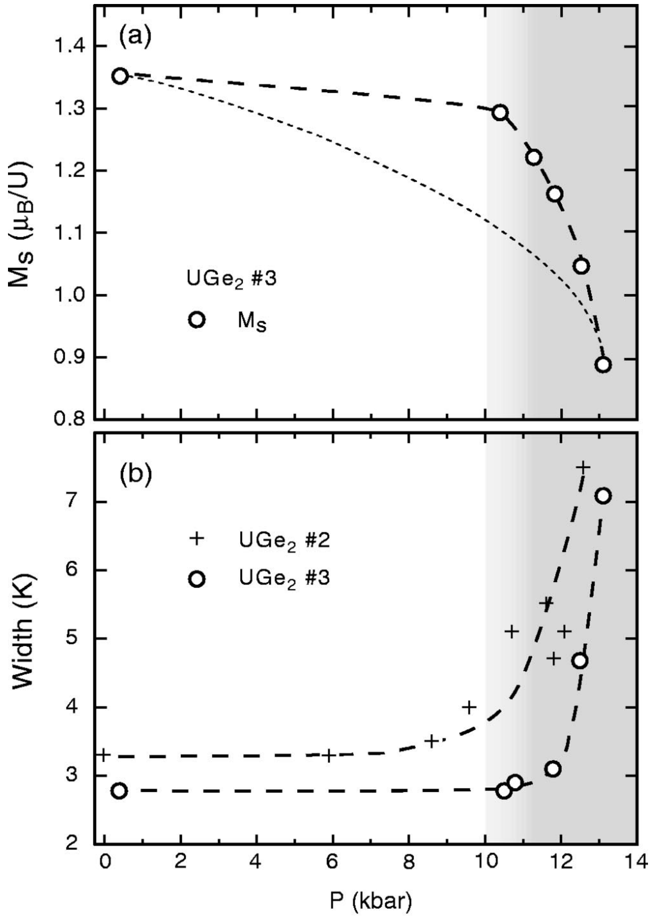

FIG. 2. (a) Pressure dependence of saturation magnetization at low temperature ($\sim$1 K), M_{S}, for the sample No. 3. (b) Pressure dependence of a full width of a half maximum of the peak in $\chi_{\mathrm{ac}}(T)$ at T_{Curie} for the samples No. 2 and No. 3. Broken lines are guides to the eyes. A dotted line denotes the equation $M_{\mathrm{S}}(P) = [T_{\mathrm{Curie}}(P)/T_{\mathrm{Curie}}(0)]^{2/3} M_{\mathrm{S}}(0)$, predicted from the SCR theory (see text). All of these quantities exhibit a slight and gradual change in the normal state, while they show a steep variation in the superconducting phase that was denoted by a shaded region. (Since the lowest temperature accessible in the present measurements, $\sim$0.35 K, is not low enough to exactly determine a phase boundary between the normal and superconducting phases, we showed the phase boundary by gradation.) These results indicate a correlation between the ferromagnetism and the superconductivity.

It was pointed out by Oomi *et al.* that a derivative of electrical resistivity vs temperature curves shows an anomaly at a characteristic temperature denoted by T'.[7] Comparing the reported data with our results, we found that these two characteristic temperatures T' and T_{M} coincide with each other. According to theoretical investigations by Watanabe and Miyake,[8] these anomalous behaviors can be explained by fluctuations of charge density waves, but it seems possible that the anomalies may be ascribed to other effects such as magnetic domain effects.[9] Further detailed study is needed to reveal an origin of the anomaly.

Measurements of $M(T)$ at external pressures give the P dependence of a saturation magnetization at low temperature ($\sim$1 K), M_{S}, the results being given in Fig. 2(a). (The measurements of the dc magnetization were made by applying the external field of 2 kOe, but we found from the magnetization process measurements that the strength of the field is small enough to estimate $M_{\rm S}$ extrapolated to zero external magnetic field.) Huxley *et al.* performed elastic neutron scattering experiments under high pressures to extract P dependence of $M_{\rm S}$. They argued a possible relationship between the Curie temperature and the saturation magnetization; the standard self-consistent-renormalization (SCR) theory predicts the relation of $T_{\rm Curie}\propto M_{\rm S}{}^{3/2}$,[3] which leads to the equation of $M_{\rm S}(P)=[T_{\rm Curie}(P)/T_{\rm Curie}(0)]^{2/3}M_{\rm S}(0)$. Here, $M_{\rm S}(P)$ and $T_{\rm Curie}(P)$ are the saturation magnetization and the Curie temperature at P, respectively, and the equation is indicated by a dotted line in Fig. 2(a). The data point at 13 kbar lies on the line, but other data points strikingly deviate from the line. Instead of such a smooth variation, it is likely that $M_{\rm S}(P)$ changes its P dependence between the normal and superconducting states; $M_{\rm S}$ indicates a slight decrease initially with increasing P, and when the system enters the superconducting phase illustrated by a shaded region, it exhibits a steep decrease. (The lowest temperature accessible in the present measurements, $\sim$0.35 K, is not low enough to exactly determine a phase boundary between the normal and superconducting phases. This is why the phase boundary is illustrated by gradation.) This result strongly suggests that the ferromagnetism is affected by the onset of superconductivity.

As pointed out above (see the inset to Fig. 1), the peak at $T_{\rm Curie}$ in $\chi_{\rm ac}(T)$ of No. 3 becomes broad and weak at high pressures. This feature is more quantitatively seen in Fig. 2(b), in which a full width of a half maximum of the peak is plotted as a function of pressure; the peak width increases at a very rapid rate in the SC phase. We note here (for No. 3) that the width at 13.1 kbar is about 50% greater than that at 12.5 kbar. Such a large increase cannot be explained by an inhomogeneous pressure distribution over the sample, because a slope of $dT_{\rm Curie}/dP$ at 13.1 kbar is only several % larger than that at 12.5 kbar ($=4.8$ K/kbar). This is also the case for No. 2, as may be seen in Fig. 2(b). Therefore, the broadening of the anomaly at high pressures is of intrinsic origin, suggesting that the FM possibly becomes inhomogeneous with increasing P in the SC state.

We plot $\chi_{\rm ac}(T)$ at around $T_{\rm SC}$ as well as around $T_{\rm Curie}$ for No. 3 and No. 2 in Fig. 3(a) and (b), respectively. A value of -1 in the vertical axis corresponds to a perfect (100%) shielding effect. However, since it was not possible to determine a demagnetizing factor for the sample because of its irregular shape, the absolute value of the magnetic susceptibility is considered to be a rough measure of the shielding effect.

First we consider the SC transition shown in Fig. 3(a) (see lower part). When P reaches 11.8 kbar, a SC onset temperature $T_{\rm onset}$ exceeds 0.7 K, as manifested by a diamagnetic susceptibility below $T_{\rm onset}$. It is to be noted that the diamagnetic susceptibility seems to saturate at a value as small as -0.5 at around 0.4 K, which implies that a volume fraction of the SC state is less than 50%. When P increases to 12.5 kbar, $T_{\rm onset}$ is likely decreased, but interestingly the diamagnetic susceptibility increases to -0.9 around 0.4 K. A similar feature is also observed in No. 2, as seen in Fig. 3(b): When P increases to 11.8 kbar, $T_{\rm onset}$ reaches a maximum value of 0.9 K, but the diamagnetic susceptibility is -0.6 even at 0.4 K. When $P=12.1$ kbar, $T_{\rm onset}$ is not changed very much, but the diamagnetic susceptibility increases up to -0.9 at 0.4 K. At 12.6 kbar, $T_{\rm onset}$ decreases, while a saturated value of the diamagnetic susceptibility at 0.4 K slightly increases. These results strongly suggest that the superconductivity does not develop over a volume of the sample at pressures near the lower critical pressure P_1 ($\sim$10 kbar), and that the superconducting volume fraction grows with increasing P.

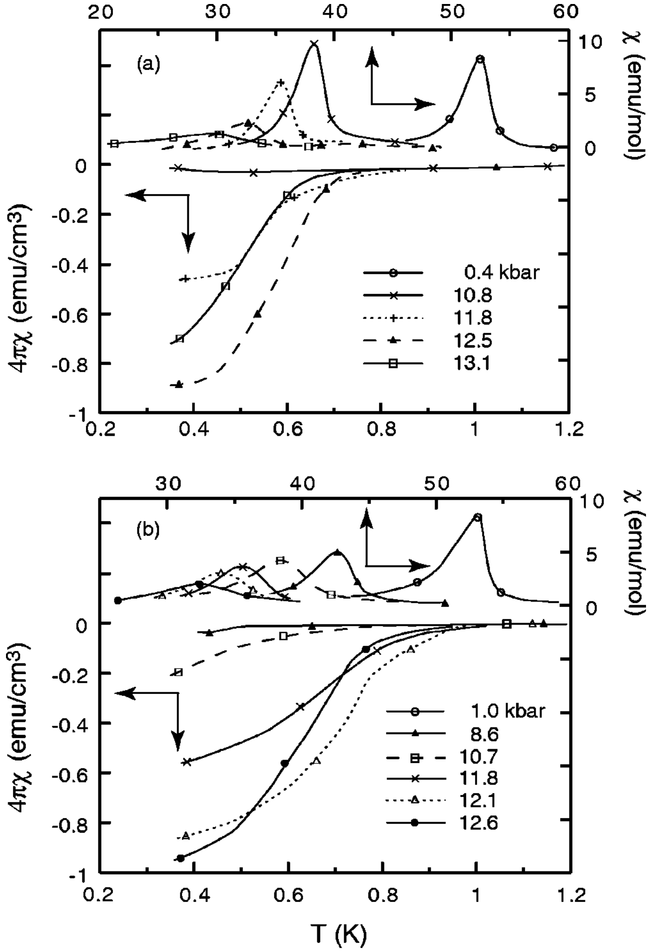

FIG. 3. (a) Temperature dependence of $\chi_{\rm ac}(T)$ around $T_{\rm Curie}$ (upper part) and around $T_{\rm SC}$ (lower) for the sample No. 3. (b) Temperature dependence of $\chi_{\rm ac}(T)$ around $T_{\rm Curie}$ (upper part) and around $T_{\rm SC}$ (lower) for the sample No. 2. These measurements were carried out by applying an ac magnetic field of $\sim$0.3 Oe (peak-to-peak value) modulated with a frequency of 100 Hz parallel to the *a* axis, and were made in decreasing temperature. Note that the anomaly due to the ferromagnetic ordering becomes substantially damped with increasing pressure, while the superconducting shielding effect becomes rather sharp with a larger saturation value.

Let us then consider the correlation between the SC and FM behaviors. The peak-anomaly due to the FM of No. 3 has a rather small width at 11.8 kbar [see Fig. 2(b)], at which the SC shielding effect is imperfect [see Fig. 3(a)]. At 12.5 kbar, the width increases by about 50%, leading to the remarkable supression of the peak, while the SC shielding effect substantially increases. The sample No. 2 shows similar features; above about 12 kbar, the peak at T_{Curie} becomes fairly broad, while the SC shielding effect at low temperatures increases with P. These results clearly show the correlation between the magnitude of the shielding effect and the width of the peak due to the FM ordering in $\chi_{\mathrm{ac}}(T)$.

We summarize our findings as follows: As P increases in normal state, M_{S} decreases gradually, and the peak anomaly in $\chi_{\mathrm{ac}}(T)$ at T_{Curie} shows only the slight increase in width. When P increases furthermore to enter the superconducting state, these P dependences are accelerated very much, in particular, the peak in $\chi_{\mathrm{ac}}(T)$ associated with the ferromagnetic transition is remarkably obscured at high pressures at which we observe the nearly perfect shielding effect. These results imply that some part of a sample remains to be in normal state when P is close to the lower boundary between the normal and superconducting phases. This is consistent with recent measurements of the specific heat at 11.3 kbar reported by Tateiwa *et al.*,[10] which showed not only a broad transition near at T_{SC} but also a very large residual electronic specific heat coefficient, $\gamma_{\mathrm{res}}=0.75\gamma_{\mathrm{n}}$, where γ_{n} is an electronic specific heat coefficient just above T_{SC}. (Since a Fermi surface of minority spin is smaller than that of the majority, γ_{res} is to be less than $0.5\gamma_{\mathrm{n}}$, even if a superconducting energy gap does not open on the minority spin Fermi surface.)

In conclusion, we suggest that the FM competes with the SC in UGe_2, which may have the following implication: In normal state the ferromagnetic ordering is homogeneous over the system. When P exceeds the lower critical pressure, a region of the SC state appears in a sample. As P increases, the volume fraction of the SC state grows, while the FM becomes spatially inhomogeneous as if the long-ranged ordered nature of FM is reduced. At present we do not talk about the homogeneity of superconductivity; let us assume that microscopic superconducting texture is formed and superconducting domains are linked to each other via weak superconducting or normal domain boundaries, and then the ac magnetic susceptibility measurements will give a nearly perfect shielding effect, as if the superconductivity would be homogeneous over the sample. It is still an open question if the SC and FM states coexist at the same part of a sample or are separated in the real space; it may be possible to suggest the possibility that in a more homogeneous sample there would be a cleaner separation between the ferromagnetism and superconductivity. Further microscopic investigations are needed to resolve these problems.

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