MAGNETIC FLUCTUATIONS IN THE ORDERED STATE OF THE FERROMAGNETIC SUPERCONDUCTOR UGe_2*

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(Received July 10, 2002)

A single crystal of the orthorhombic UGe_2 has been studied by magnetic (up to 5 T) and magnetotransport (up to 8 T) measurements. It was found that at low fields there exists a domain structure which is reconstructed to a monodomain one in fields higher than 0.1 T. The electrical resistivity has appeared to be highly anisotropic with $\rho_b \gg \rho_c > \rho_a$. For all three crystallographic directions the Fermi liquid equation is followed below about 8 K. Also the transverse magnetoresistivity (TMR) is anisotropic. At low temperatures the TMR for all three principal directions of the unit cell is positive and the values at 4.2 K and 8 T are in the following proportions 1:4:2 for TMR measured along the a-, b- and c-axes, respectively. At temperatures above about 15 (b-axis) and 25 K (a- and c-axes) the TMR becomes negative with a distinct minimum at T_C for the latter two directions. However, the most spectacular behaviour is observed when $j||b$ and $B||a$. The TMR goes through very broad negative minimum achieving as high value as –40% at T=27 K, thus close to so-called characteristic temperature T^* (= 30 K), that is established from the maximum in the temperature derivative of the resistivity. So far the effect in TMR can be considered as the most distinct manifestation of the existence of magnetic fluctuations in UGe_2 taking place at temperatures at around $1/2T_C$, without traces of a similar manifestation at T_C. The observed giant effect in magnetoresistivity can support the idea of the proximity of the system to a transition into the coupled CDW/SDW type ordering which probably takes place when applying pressure.

PACS numbers: 72.15.Gd, 75.60.–d, 75.30.Cr , 71.45.Le

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

1. Introduction

In recent years, superconductivity (SC) under pressure has been observed in some antiferromagnets, like $CePd_2Si_2$, $CeIn_3$ and others. For such a case of unconventional SC arising in these and other heavy fermion compounds, the charge carriers are claimed to be coupled in pairs by magnetic interactions [1]. This means that magnetic fluctuations replace vibrations in the formation of Cooper pairs. Furthermore, it was shown that such type of pairing is most robust in quasi two-dimensional (2D) systems [2].

The recent discovery of SC in the UGe_2 ferromagnet (T_C=53 K) under pressure [3] was quite surprising, because the huge inner molecular field here ($H_{hf} \approx 240(10)$ T at 5.3 K [4]) is not cancelled out, like in the case of antiferromagnets mentioned above. Under such a strong internal field it is claimed that only the nonunitary triplet state can survive [5].

The most important difference with respect to the above antiferromagnets is that the SC in UGe_2 is enclosed into the ferromagnetic phase and disappears together with the ferromagnetism at the critical pressure p_C=16 kbar, where T_C=0 K, *i.e.* at the quantum critical point (QCP) [3]. Although the possibility of a p-wave coupling has been predicted in itinerant ferromagnets [6, 7] some time ago, the appearance of SC was rather associated with the QCP. The absence of SC also in the paramagnetic phase of UGe_2, i.e. for $p > p_C$ is related to the first order transition taking place at p_C [8].

It is commonly established, that the most important feature in creation of SC in UGe_2, mediated by magnetic fluctuations, is just connected with the observation of a broad anomaly in the temperature derivative of the resistivity having a maximum at so-called characteristic temperature T^* first reported by Oomi *et al.* [9, 10] This temperature taken at ambient pressure is about 30 K and reaches zero at p_C^*=12 kbar, where T^* vanishes and T_{SC} becomes the highest (0.8 K) [3, 8]. At the same time T_C=32 K and μ_s=1 μ_B. This transition into such unconventional SC in the presence of ferromagnetism appears to be of second order in contrast to that in p_C and though there is no experimental proof, the new mechanism driving the system into this kind of SC has recently been discussed [3, 8] and considered theoretically [11, 12] on the basis of the appearance of coupled CDW–SDW critical fluctuations at $T^*(p)$ phase boundary. Until now the nature of this characteristic temperature has not been well recognized and therefore it has become a matter of intensive debates in the literature.

In our previous publication [13] we have pointed out that the peak in the $d\rho(T)/dT$ curve observed at 30 K at ambient pressure reflects the fact that the $\rho(T)$ dependence goes through an inflection point at T_i (= 30 K). Such a point in the case of the non-magnetically ordered spin fluctuators, like UAl_2 and UPt_3 [14] is associated with the spin-fluctuation temperature T_{sf}. A

similar standpoint has been also presented in Ref. [15]. Accordingly, there is also claimed that the anomaly at $T_{\rm i}$ should be rather related to a crossover boundary than to a phase transition at ambient pressure. However, at higher pressures, but below $p_{\rm C}^{*}$, a distinct change of a slope in the temperature dependence of the resistivity, apart from that at $T_{\rm C}$ [8,16] or even a jump in the magnetization [17,18] takes place, both of which may signal the existence of a real transition boundary, *e.g.* into the CDW/SDW mode within the ferromagnetic state [12].

UGe_2 crystallizes in the orthorhombic structure of *Cmmm* space group ($ZrGa_2$-type) with large b/a and b/c ratios of 3.76 and 3.68, respectively. In this structure the uranium atoms are arranged in zigzag chains with shortest U–U distance of 0.382 nm, that run along the [100] direction [19,20]. These chains are separated out by the Ge atoms (the coordination number is 10) located in the *ab* and *ac* planes.

The magnetization [21,22] and neutron diffraction [8,20,23] experiments made on UGe_2 have revealed the Ising type anisotropy in this compound with the magnetic moments amounting to 1.4 $\mu_{\rm B}$ arranged collinearly to the shortest axis *a*.

The de Haas–van Alphen (dHvA) experiment indicated that the main Fermi surface (FS) is cylindrical with an axis along the *b*-axis [24–26]. For such a pseudo- 2D-topology of the FS one can thus expect an effect of the nesting in the majority-spin band, *i.e.* favoring the occurrence of CDW/SDW modes, which leads to an opening a gap in some part of the FS [11,12,25].

In order to address the question of the presence of ferromagnetic fluctuations associated for example with the formation of the CDW–SDW modes at T^{*} (p=0) we have studied here the field and temperature dependent transverse magnetoresistivity (TMR), defined as $\Delta\rho/\rho = [\rho(B) - \rho(0)]/\rho(0)$.

Previously the transverse magnetoresistivity was measured for a UGe_2 single crystal along three principal directions by Onuki *et al.* [27] up to B=15 T but only at 0.5 K. They found positive TMR with a $B^{1.6}$ dependence for the current j along the *c* and *b* axes and its saturation along the *a*-axis. From the dHvA experiment they detected carriers with heavy cyclotron masses up to 24.5 μ_0. The large cyclotron masses observed in this compound were considered by these authors as being caused by spin fluctuations.

We first measured MR for a polycrystalline sample of UGe_2 but in the wide temperature range 4.2–70 K in fields up to 8 T [13]. However at higher temperatures the MR becomes gradually negative as for a normal ferromagnet, reaching a maximum in the $\Delta\rho/\rho$ value as high as –27% at 26 K and 8 T. Thus this giant anomaly occurs at the temperature rather close than exact at T^{*}, while the MR at $T_{\rm C}$ was found to be almost zero. It appears that this kind of behaviour is completely strange for a normal ferromag-

net. Usually for such a material below T_C, the electron scattering gradually diminishes with no further contribution to the total MR at temperatures close to 0 K due to the uniform aligning of magnetic moments along one direction [28]. Therefore it seems that the real magnetic properties of UGe_2 have not been so far completely recognized in view of giant electron scatterings observed well below T_C. Thus, this marked MR effect occurring at T=26 K, *i.e.* only slightly lower than T_i, strongly indicates that the coupled spin and charge fluctuations predicted by Watanabe and Miyake [12] can really develop even deep within the ferromagnetic phase and in consequence they may play an essential role in the mechanism of SC formation in UGe_2 under pressure. This temperature, now called the spin-fluctuation temperature T_{sf}, seems to be slightly lower than T_i ($\equiv T^*$), also indicated in earlier studies of thermal expansion of single crystalline UGe_2 [9], where anomalies in the thermal expansion coefficient occur rather around T=25 K than right at T_i. It should be also mentioned that the authors of Ref. [12] have pointed out that the onset of a CDW ordering in the majority-spin band is connected with the observed anomalously large lattice contribution to the heat capacity at ambient pressure with a broad maximum also just around T=25 K.

In this paper some detailed studies have been made on a UGe_2 single crystal, which allow one to demonstrate the presence of a strong anisotropy in the transverse MR in this compound and confirm the occurrence of a giant effect in the magnetoresistivity at $T \approx T_C/2$.

2. Experiment

The obtained single crystals by Czochralski method were checked by X-ray and electron microprobe analyses. Both give evidence of single phase character of the material used for further measurements. The lattice parameters were: a=0.4018(1) nm, b=1.5083(3) nm and c=0.4098(1) nm. For magnetic and electrical measurements a larger irregular crystal was cut into nearly rectangular parallelepipeds having the longest axis along a given direction of measurements. Typical sample dimensions of $0.5\times0.5\times4$ mm^3 were used for the electrical measurements. The electrical resistivity measurements were done by using a standard DC-four probe method with an accuracy of better than 10^{-6}. Electrical contact to the sample was made by In-soldering (current) and spark attaching (voltage). MR was measured in fields up to 8 T. Three different samples were studied depending on the kind of measurements. We report in this work the results obtained on one of our samples, though the data were verified on three different samples. Magnetization measurements were made by a SQUID magnetometer from 1.7 to 300 K in magnetic fields up to 5 T.

3. Results and discussion

The magnetization of UGe_2 measured for three principal crystallographic directions were done many years ago [21, 22] and our results confirm completely the existing data at 4.2 K. In addition, the magnetization measurements on the UGe_2 crystal oriented along the easy a axis were made for the first time in the wide ranges of temperatures and applied magnetic fields. Fig. 1(a) shows the results obtained for the magnetic fields up to 5 T and for a number of fixed temperatures from 1.7 to 80 K, while Fig. 1(b) displays the results obtained at temperatures up to slightly above the Curie temperature (T_C) and at fixed magnetic fields (B) between 0.005 to 1 T. From these figures it is clear that the magnetization becomes linear in applied magnetic fields at temperatures higher than about 30 K above T_C and shows very distinct anisotropy in zero-field cooling runs performed for several fixed fields. It gives some idea about the existence of some short range magnetic order above T_C on the one hand and a domain effect on the other. Hence, one sees that the monodomain behaviour can be achieved in applied fields as high as about 1 T.

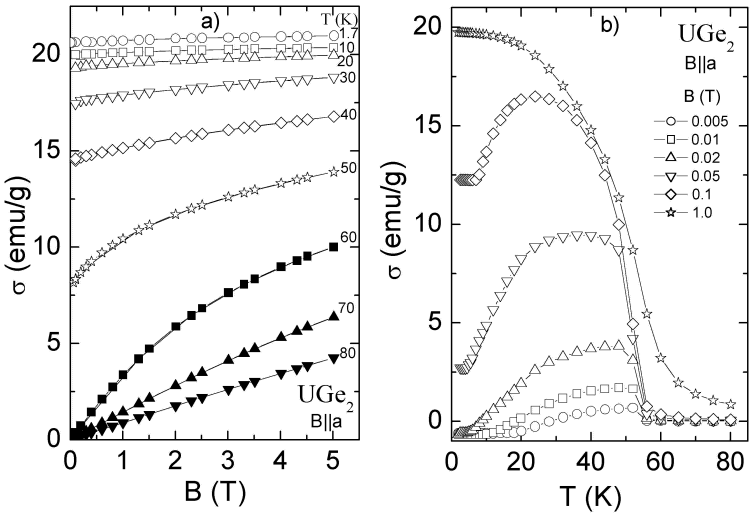

Fig. 1. Magnetization as a function of (a) applied magnetic field and (b) temperature, both measured along the easy axis a.

Fig. 2 displays the electrical resistivity of UGe_2 against temperature with the current j along the a-, b- and c-axes. The residual resistivity ratios (RRR) appear to be almost one order of magnitude higher than those reported in the literature, for example for the samples used in studies of the dHvA effect (see *e.g.* Ref. 24). As in the earlier works [22], the resistiv-

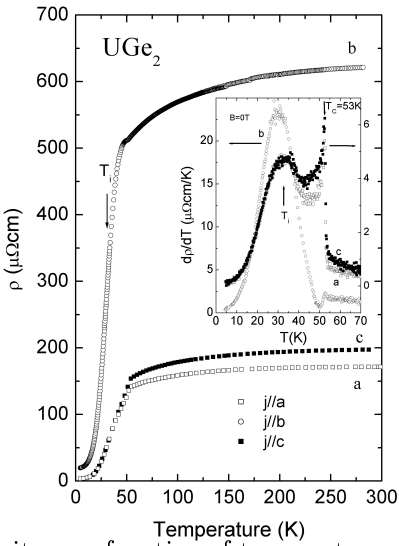

Fig. 2. Electrical resistivity as a function of temperature measured along the three main crystallographic axes : a, b and c. The inset shows the temperature derivative of the resistivity as a function of temperature for these three axes.

ity below T_{C} shows steep decrease due to vanishing spin-disorder scattering with decreasing temperature. In all the cases the $\rho(T)$ curve goes through an inflection point T_{i} as also shown in the inset of Fig. 2 in the form of a maximum in the temperature derivative of the resistivity. However, if for the a- and c- directions this derivative shows very sharp peaks at T_{C}, while that for the b-axis it is rather small and diffused. Also for this axis, a hump in the resistivity takes place just below T_{C}, as was reported earlier by Onuki *et al.* [22]. It should be noted that the overall values of the resistivity along the b-axis for the sample considered here are considerably higher than those reported by these authors, while another sample which has been also measured by us (not shown) behaved according to the results of Ref. [22]. The corresponding ρ_b/ρ_a ratios at room temperature are 1.76 [22] and 3.65 [this work]. At present it is difficult to decide if this discrepancy arises from a different alignment of crystals or whether it is caused by faults in stacking along the b-axis. The $\rho(T)$ dependence above T_{C} in all the cases shows a negative curvature characteristic for the $\rho(T)$ curves encountered for spin-fluctuators.

At low temperatures the resistivity follows up to about 8 K the Fermi liquid equation: $\rho(T) = \rho_0 + AT^2$ for three principal directions of orthorhombic UGe_2 with the A values of about 0.01 $\mu\Omega\mathrm{cm/K}^2$ for j along the a-and c-axes, while for the hard b-direction this value is about 0.05 $\mu\Omega\mathrm{cm/K}^2$. These values are of the same order of magnitudes as those reported in the literature for the polycrystalline samples [15] as well as for single crystals [10, 16].

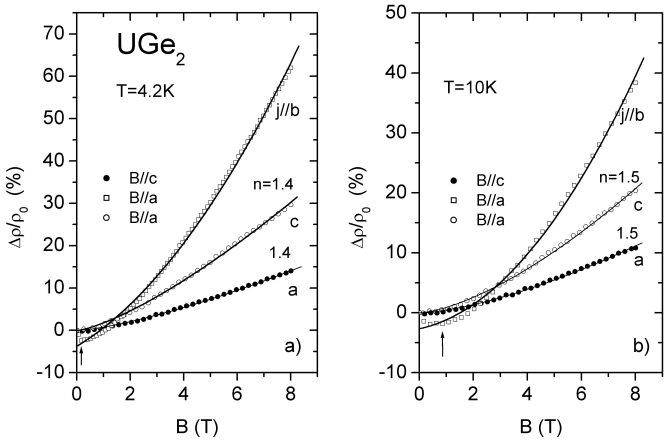

Fig. 3. Transverse magnetoresistivity (TMR) as a function of magnetic field applied along three main crystallographic axes a, b and c at temperatures (a) 4.2 K and (b) 10 K. The solid lines for the a and c axes represent a simple AB^n fit (n is displayed), while along the b axis a more complex equation has to be used : $\Delta\rho/\rho = -a + bB + cB^2$.

Fig. 3 displays the magnetoresistivity measured at 4.2 and 10 K for j directed along three main axes a, b and c as a function of magnetic field applied always perpendicular to a given j direction. As seen, the TMR is large and positive emphasizing first of all the fact that the Fermi surface (FS) effect is important at low temperatures, especially when the current is parallel to the hard b-axis for which $(\Delta\rho/\rho)_b$ measured at B=8 T is equal to 62 and 40% at 4.2 K and 10 K, respectively. Thus along this axis the largest electron orbits have been found in the dHvA effect measurements [24]. The least-squares fits to the TMR data along the a- and c-axes at these two temperatures show the expected AB^n behaviour with $n \approx 1.5(1)$. However, due to the negative $(\Delta\rho/\rho)_b$ values persisting almost up to 2 T another equation has been used:

$$(\Delta\rho/\rho)_b = -a + bB + cB^2. \tag{1}$$

Subsequently, the ratios of the MR magnitudes found at 8 T for j along the a-, b- and c-directions are almost the same at these two temperatures amounting to about 1:4:2 which gives a scale of the anisotropy in the magnetotransport properties of UGe_2. In general these data are in accordance with the first measurements of Onuki *et al.* [27] at 0.5 K but only where $j||b$ and $B||a$ and c. In contrast to our results obtained at 4.2 K, these authors claimed a saturation behaviour at 0.5 K in higher fields for the cases where $j||c$ or a and $B||a$ or c, signalling the presence of open electron orbits. This

difference in the results may arise from the rapid change of the Fermi surface topology with temperature. This seems to be consistent also with the TMR results of Oomi *et al.* [29] obtained for $j||a$ and $B||b$ at 4.2 K and 5 T. These authors present the $(\Delta\rho/\rho)_a$ *vs.* B curves also without any apparent tendency to saturation.

In Figs. 4a and 4b and their insets we display the electrical resistivity and its temperature derivative as a function of temperature, respectively, where both these dependences were estimated at B=0 and 8 T for two cases a) $j||a$, $B||b$ and b) $j||b$, $B||a$. Especially the temperature variations of the corresponding derivatives are different. If for the former condition $T_{\rm i}$ is not changed in contrast to $T_{\rm C}$, then for the second condition both these characteristic temperatures are markedly shifted to higher temperatures by about 10 K. As we will see below, this behaviour is opposite to the temperature of the negative minimum at $T_{\rm sf}$ found in the $(\Delta\rho/\rho)_b$ *vs.* T curve which at 8 T is lower than $T_{\rm i}(\equiv T^*)$ determined at B=0 T. This may indicate that the real characteristic temperature T^* having been determined up to now from the maximum in the temperature derivative of the resistivity is not exactly that which we expect to be a crossover or a transition one.

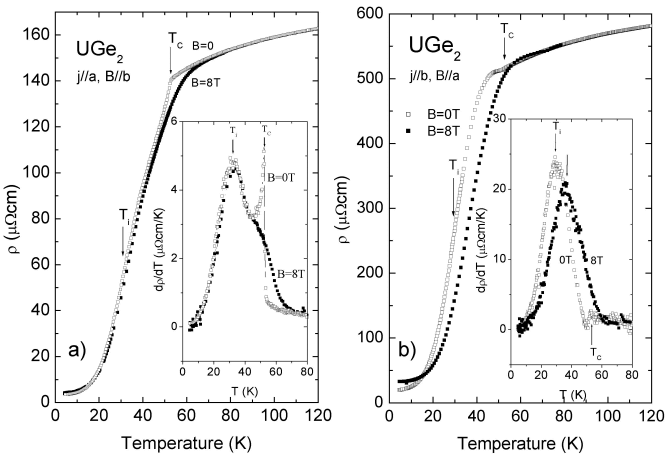

Fig. 4. Electrical resistivity as a function of temperature measured at B=0 and 8 T with the current j along (a) easy axis a and (b) the hard axis b. The insets show the temperature derivative of the resistivity against temperature.

In Fig. 5(a) the field dependence of $(\Delta\rho/\rho)_a$ is demonstrated where $B||b$ (the case studied in Ref. [29]). Like the situation which we have analyzed above, there is no a simple AB^n behaviour, but more complex one which may be described *e.g.* by Eq. (1) for two temperatures 4.2 and 10 K. In turn, Fig. 5(b) presents a large difference in the MR behaviour when the temperature $(\Delta\rho/\rho)_a$ dependences are measured under magnetic fields B

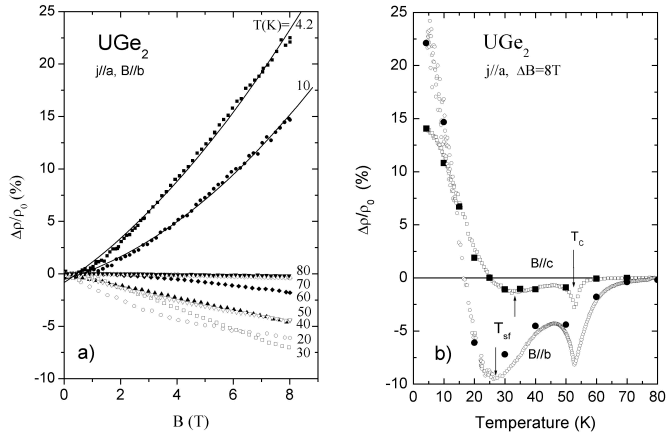

Fig. 5. Transverse magnetoresistivity measured as a function (a) of applied magnetic field for $j||a$ and $B||b$ and (b) of temperature for $j||a$ and $B||c$ as well as for $j||a$ and $B||b$. The large closed circles and squares are the values taken at 8 T from the relevant $\Delta\rho/\rho$ *vs.* B dependencies measured at various temperatures.

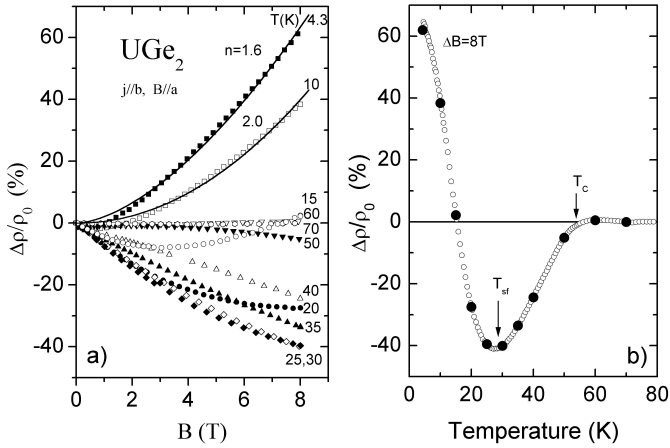

Fig. 6. The TMR measured for the current $j||b$ and field $B||a$ where (a) $\Delta\rho/\rho$ is taken at various temperatures and in fields up to 8 T and (b) $\Delta\rho/\rho$ is taken at an isofield of 8 T between 4.2 and 80 K (small open circles). Large filled circles are data taken at 8 T at various temperatures as shown in panel (a).

directed either along the c- or b-axes. For the latter case both $T_{\rm sf}$ and $T_{\rm C}$ are very well observed as a very broaded and a sharper negative peak, respectively.

However, the most interesting case is presented in Fig. 6, where (a) the field- and (b) the temperature-variations of $(\Delta\rho/\rho)_b$ are given. One can

see very distinctly from Fig. 6(a) that at about 15 K, where the change in the sight of MR takes place (see Fig. 6(b)), and at temperatures somewhat higher, the $(\Delta\rho/\rho)_b$ *vs.* B variation shows the presence of some competitive contributions one positive and another one negative. The latter one increases with increasing temperature showing a typical ferromagnetic-like tendency of MR to saturate. Subsequently, the curvature of this function at temperatures above 30 K becomes opposite, rather reminiscent of those for Kondo-like or paramagnetic-like variations of MR with the field, despite the presence of ferromagnetic order. Undoubtedly, the most intriguing result is that the MR exhibits a value as high as –40% at $T_{\rm sf}$ and almost 0% at $T_{\rm C}$ when the current is along the hard magnetization direction and the field is applied along the easy magnetization direction. This finding points to the presence of strong magnetic fluctuations along this hard direction at the temperature just midway up to the Curie temperature, which are being frozen out in an applying magnetic field. Finally, Fig. 7 demonstrates the reduced TMR measured for several magnetic fields as a function of temperature, keeping the same conditions as those presented in Fig. 6. It is clear from this figure that TMR measured at several temperatures in the constant magnetic field of 1 T is negative for the entire range of temperatures between 4.2–70 K. However, with increasing magnetic field the MR values at low temperatures increase rapidly to large positive values as is expected

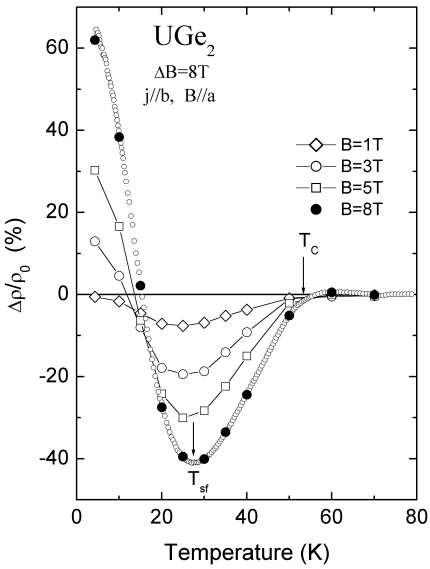

Fig. 7. TMR measured in different fields as a function of temperature. The curve taken at 8 T is the same as in Fig. 6.

from the AB^n dependence, while near a temperature T_{sf} (= 25(2) K) a deep negative minimum is formed with a very small shift to higher temperature, but simultaneously a negligible change is observed at around T_C.

4. Conclusions

Recently, we have discovered unusual temperature dependence of the transverse magnetoresistivity of a polycrystalline sample of UGe_2 [13]. At present we have extended these studies on single-crystalline samples of this compound. So far such studies for this recently discovered ferromagnetic superconductor have been explored at very low temperature as 0.5 K [27]. The TMR for UGe_2 at low temperatures exhibits nearly quadratic field dependencies for the current directed along the three principal axes, but with a high anisotropy in their magnitudes.

At ambient pressure the largest effect in TMR was observed in the case of the current j parallel to the hard magnetization direction b, being simultaneously the most elongated axis of the unit cell of UGe_2, and with magnetic field applied along the easy a axis. This effect amounting to about –40% appeared at T=26 K at 8 T which is a half of the Curie temperature value, while at T_C itself the TMR is close to zero. Thus this novel feature is probably associated with the appearance of strong magnetic fluctuations signalling that the system is in close proximity of a phase transition, what probably happens when applying external pressure. The conclusion of fluctuations instead of a transition is also indicated by a broad excess-anomaly in the heat capacity existing around T_{sf} [12]. These fluctuations probably coexist with magnetic excitations in the ferromagnetic order. It strongly suggests that thus the low-energy fluctuations associated with p_C^* could play a significant role in formation of SC in UGe_2 under pressure.

The distinct anisotropy for the temperature dependencies of thermopower $S(T)$ and thermal conductivity $\kappa(T)$ depending on the main crystallographic directions has also been observed [30]. In particular for the case of the former dependence, $S(T)$ goes through a distinct positive minimum below T^* (T_{min}=25(1) K) for the a and c directions and a negative minimum at the same temperature for the b direction. For all three cases there was observed a rapid drop in $S(T)$ below T_C.

This work was supported by the Polish State Committee for Scientific Research (KBN), project No. 2PO3B 150 17. The author thanks Dr. J. Stępień-Damm for X-ray measurements and D. Badurski and R. Gorzelniak for their technical assistance in this work.

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