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f-electron Dualism as a Reason of Superconductivity in Ferromagnetic UGe_2

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Magnetization and electrical resistivity measurements in the wide temperature and field ranges were performed on single crystals of UGe_2. The presence of extremely large anisotropy in the temperature dependence of the susceptibility and magnetization has confirmed the previously published results. The dependence of spontaneous magnetization, σ_s, on temperature has been inferred from the Arrot plot and compared to that determined from neutron diffraction data. The earlier transversal and present longitudinal results on the magnetoresistivity are discussed in the terms of possible f-electron *dualism* in UGe_2 and its connection with the occurrence of superconductivity under pressure in this compound.

Key words : f-electron magnetism, magnetoelectron resistivity, superconductivity

1. Introduction

The discovery of superconductivity (SC) under pressure in the ferromagnetic UGe_2 (T_C=53 K and $\mu_S \approx 1.4\ \mu_B$ at ambient pressure) [1] was associated with the observation of a broad anomaly in the temperature derivative of the resistivity having a maximum at so called the characteristic temperature T^*(≈30 K), first reported by Oomi [2]. T^* was also clearly detected in the coefficient of volume thermal expansion α_V [3]. This coefficient first decreases discontinuously at T_C and then slowly increases with cooling exhibiting a small dip feature at $T^* \approx 25$ K, suggesting a somehow phase transition in the ferromagnetic phase. As a reason of some scattering in the observed T^* values, the authors of Ref. [3] have given the fact of using different single-crystalline samples in the electrical and thermal expansion measurements.

This temperature at ambient pressure is around 30 K and just reaches zero at the critical pressure $p_C^* \approx 12$ kbar, where T_{SC} becomes the highest (0.8 K) [1]. At the same time, $T_C = 32$ K and $\mu_S = 1\ \mu_B$, and the ferromagnetism (FM) disappears at QCP, where critical pressure $p_C \approx 16$ kbar. Until now the nature of the characteristic temperature T* has been a matter of intensive debates in the literature. Most of the works devoting to the coexistence of SC and FM in UGe_2 have been focused so far on the electronic and magnetic properties of this compound under pressure. It is natural to think, however, that the proceeding reason of the occurrence of SC in UGe_2 under pressure lies first of all in detailed recognizing these properties at ambient pressure.

Recently, we have discovered around T^* an unusual temperature dependence of the transverse magnetoresistivity (TMR), first on the poly- [4] and then on single-crystalline [5] UGe_2 samples. The TMR measured for a UGe_2 single crystal along the three main axes was found to be highly anisotropic. At low temperature the TMR for the a, b, and c axes is positive and its magnitudes at 4.2 K and 8 T are in the following proportions 1:4:2, respectively. It should be mentioned here that in the orthorhombic crystal structure of $ZrGa_2$-type in which UGe_2 crystallizes, the lattice parameter b/a ratio is as large as 3.75. This clearly shows that the smallest effect is for the easy axis a and the largest one is for the most hard direction b. At temperatures above about 15 K (b-axis) and 25 K (a- and c-axis) the TMR becomes negative with a distinct minimum at T_C, as one would expect for a ferromagnet, but only for the latter two directions. However, the most spectacular behaviour is observed when the current j//b and the magnetic field B//a. Thus for this configuration the TMR goes through a very broad negative minimum where it achieves as high value as –40% at T=27 K, i.e. at the temperature close to the

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characteristic temperature T^*. Therefore, this effect in TMR can be considered as the most distinct and peculiar manifestation of the existing in UGe_2 of some kind of strong magnetic fluctuations taking place in the ferromagnetic order just at the temperature $T_{sf} \approx T_C/2$ being close to T^*, but surprisingly without a trace manifestation at T_C.

In this work more detailed data are given on the magnetic and electrical properties of UGe_2 determined at ambient pressure on single-crystalline samples.

2. Experimental and Results

Two single crystals of UGe_2 denoted I and II were grown by a Czochralski method, as described earlier in Ref. 5. In this reference also the characterization of the sample I is given.

We synthesized also a polycrystalline sample of $ThGe_2$ by melting the components in an arc-furnace under argon atmosphere and followed annealing under vacuum at 800 °C for 4 weeks. X-ray examination revealed a single phase of the $ZrSi_2$-type (s.g. Cmmm) with the lattice parameters: a = 1.6623(2), b = 0.4019(4) and c = 0.4144(4) nm. Any attempt to obtain the isostructural to UGe_2 phase of $ThGe_2$, i.e. of the $ZrGa_2$ type (s.g. Cmcm) has failed. The latter phase was previously reported by Brown [6] in his crystal structure refinement of both types. Both these type structures, however, are closely related to each other and can be derived by a simple crystallographic translation.

The measurements of susceptibility, magnetization and electrical resistivity were made on both as-grown single crystals of UGe_2. The magnetization measurements were carried out in magnetic fields up to 5 T in the temperature range 1.9~300 K using a SQUID magnetometer. The electrical resistivity measurements in zero (up to 300 K) and magnetic-fields up to 8 T (up to 90 K) were made by the dc-four probe method. The resistivity measurements of $ThGe_2$ were performed on a rectangular parallelepiped cut off from a bulk sample.

2.1. Magnetic susceptibility

Fig. 1 displays the temperature dependence of the reciprocal paramagnetic susceptibility for the field oriented along the a-, b- and c- axes. In accordance with previous such studies [7, 8], a large anisotropy in these dependencies is well apparent. Most probably that the observed anisotropy in the susceptibility behaviour, where $\chi_a >> \chi_c > \chi_b$, is clear evidence that essentially the localized 5f electron states exist in UGe_2 and the crystal electric field (CEF) effects is mainly responsible for this

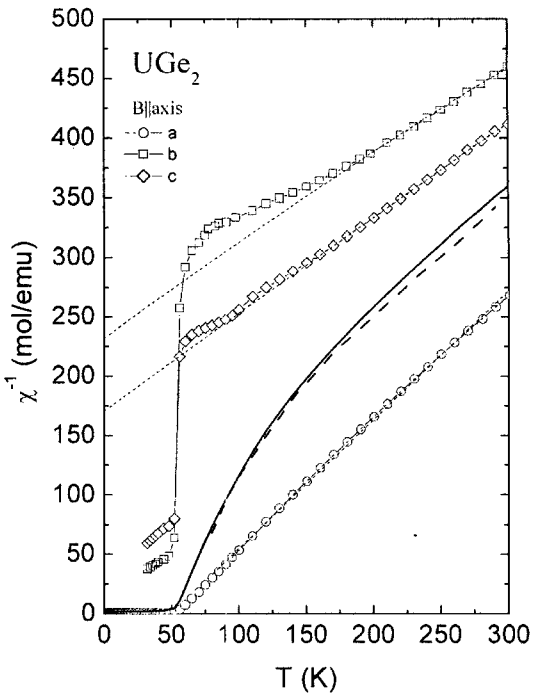

Fig. 1. Reverse molar magnetic susceptibility measured for magnetic field applied along three main crystallographic axes a, b and c. In addition the calculated curves: $\chi_{av}^{-1}(T)$ (solid line) and $\chi^{-1}{}_{poly}$ (T) (dashed line) taken from Ref. 7 are also given.

feature. The obtained magnetic characteristics by fitting the susceptibility to the modified Curie-Weiss law are presented in Table 1. There are both small and large differences compared with the previous not so precise data [7, 8] as those presented here. Menovsky *et al.* [7] found that the susceptibility measured in fields applied along the easy axis a (χ_a) is well defined by a Curie-Weiss law with $\mu_{eff} = 2.67\ \mu_B$ and $\theta_{CW} = -53$ K, while Onuki *et al.* [8] on the basis of the Curie-Weiss law fitting to their susceptibility data, but above 200 K, have estimated the effective numbers of Bohr magnetons (and the paramagnetic Curie temperatures) to be 3.0 μ_B (0 K), 3.2 μ_B (−270 K) and 3.1 μ_B (−190 K), respectively. In Fig. 1 there are also shown the temperature variations of an average susceptibility, $\chi_{av} = \chi_a + \chi_b + \chi_c/3$, compared to the reported χ_{poly} results [7], obtained on the polycrystalline sample. As seen, the $\chi_{av}^{-1}(T)$ and $\chi^{-1}{}_{poly}$ (T) curves coincide. No such a good agreement exists with respect to

Table 1. The MCW parameters for UGe_2 single crystal I

Axes	ρ_0 (10^{-3} emu/mol)	T_p (K)	μ_{eff} (μ_B)
a	0.43	56	2.55
b	0.38	-254	2.83
c	0.15	-199	3.02

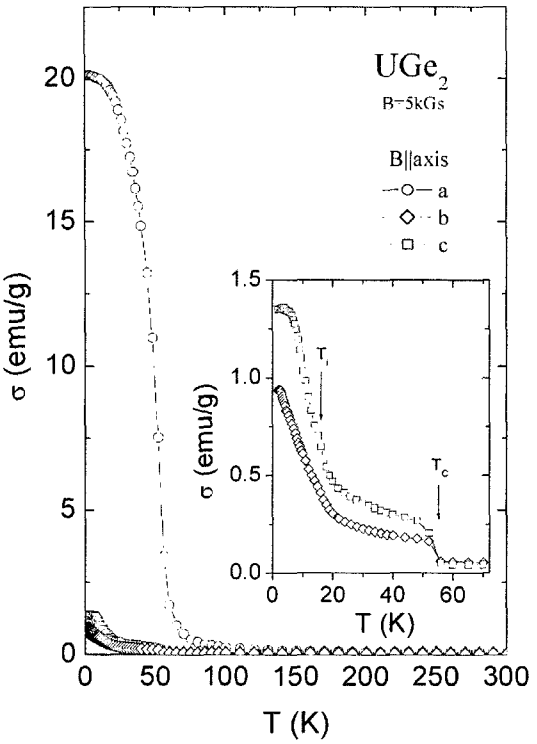

Fig. 2. Magnetization σ_a as a function of temperature. This function for σ_b and σ_c are shown in the inset.

the data of Ref. 8, probably cased by some preferential effect in grain orientation. It is clear that the strong curvature in χ^{-1}_{poly} versus T arises just from the large anisotropy in the susceptibilities, measured along main crystallographic directions.

Considering the temperature variation of the reverse susceptibilities with fields applied along the b- and c-hard directions, one notes the upward deviations in $\chi_i^{-1}(T)$ for both cases (i = c and b) below about 200 K, which might signal the presence of the CEF splitting with singlets as the ground and excited states corresponding to the $5f^2$ electronic configuration of the uranium atoms in UGe_2. Hence the FM in UGe_2 and so huge anisotropy probably are of induced type [9].

In the $ZrGa_2$-type structure the central U atom is surrounded by the 8 nearest germanium atoms which form a slightly distorted antiprismat UGe_8 yielding strongly an axial CEF potential on the central magnetic atom just along the b-axis. As is well known, for the regular antiprismatic configuration having D_{4d} point symmetry around the U^{4+} ion ($^3H_4$-term) one would deal with four non-Kramers doublets: $|\pm4>$, $|\pm3>$, $|\pm2>$ and $|\pm1>$ and one singlet $|0>$. One of them may be a ground state depending on the c/a ratio (here the b/a ratio because a~c) and the remaining states form the excited states (see for example Ref. 10). The parametrization of the crystal field

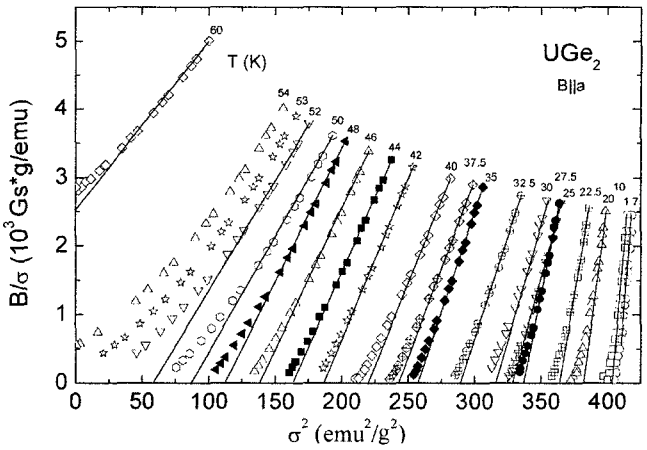

Fig. 3. The Arrot plot taken along the easy magnetization axis a.

potential and the splitting diagrams for the lower symmetries than D_{4d}, yielding only CEF singlets, where one of scheme could be adequate for the case of UGe_2, has been reported, e.g. in Ref. 11 in Fig. 3. Closer inspection of the antiprismat indicates, that the distances: U-4Ge(1), U-2Ge(2) and U-2Ge(3) are close to one another and are as follows: 0.297, 0.295 and 0.292 nm, respectively. The two other distances U-2Ge(1) in the case of UGe_{10} configuration equaling to 0.323 nm, seem to be too large, and they should not be taken into account..

2.2. Magnetization

The magnetization curves measurement performed at 1.9 K along main crystallographic directions (not shown here) gave very similar results to those already reported in the literature [7, 8]. These measurements have confirmed that the direction of an easy magnetization is the shortest axis (a-axis) and that the value of the ferromagnetic moment along this axis is about 1.4 μ_B. The magnetization curves taken for this axis at temperatures between 1.7 and 80 K together with those showing temperature dependence of magnetization, measured in fields from 50 Gs to 10 kGs were presented elsewhere [5]. Here, there is demonstrated in Fig. 2 the thermal dependence of the magnetization taken at 5 kGs along three main crystallographic directions. This figure illustrates first of all the fact how small values of the magnetic moments are reached at 1.9 K for the b- and c-hard directions (0.07 and 0.096 μ_B, respectively) in relation to the value of 1.4 μ_B found along the easy axis. Thus, this gives also a scale of huge anisotropy. In the inset of Fig. 2 the temperature variations of the magnetization along the hard b- and c-axes are shown. It appears, that these dependencies are not of a Brillouin curve type and differ in their shape from that measured along the easy axis, which is typical

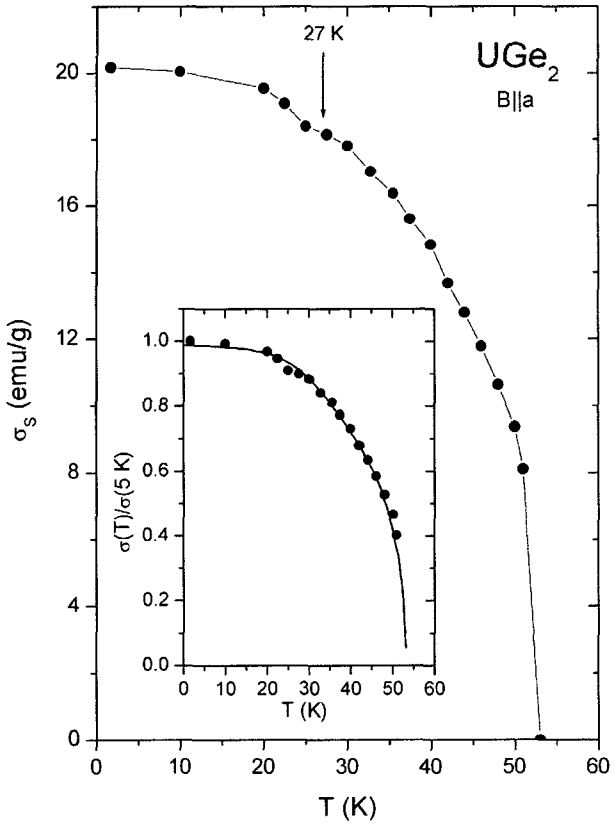

Fig. 4. The extracted from the Arrot plot (Fig. 3) the spontaneous magnetization s_s as a function of temperature (solid circles). The inset shows for the reduced $\sigma_s(T)/\sigma_s(5\ K)$ magnetization (solid circles) compared to that determined in the neutron diffraction measurement, taken from Ref. 17 (solid line), both are plotted as a function of temperature.

for a ferromagnet. Namely, for the b- and c-axes the s versus T curves show an inflection point at about 15 K followed by a long tail up to T_C.

Magnetization data in a wide temperature range from 1.9 K to 60 K are presented in Fig. 3 in fashion of so called "Arrot plot", i.e. σ^2 versus B/σ. The extrapolation of high field B/s values to zero yields the square spontaneous magnetization σ_s^2. Then the σ_s values were inferred from the above extrapolation and have been plotted against the temperature in Fig. 4. Although it concerns practically the only one extracted point, a small depression in the vicinity of T^* is seen. Furthermore, their reduced values $\sigma_s(T)/\sigma_s(0)$ are also compared in the inset of this figure to a similar curve obtained from the neutron diffraction data [1], An agreement between these two experiments is quite good. However, it is interesting to note that such a simple extrapolation used at lower temperatures fails at higher temperatures to get $T_C \approx 53$ K as expected. Earlier such a plot but for only several temperatures around T_C was presented by Menovsky *et al.*

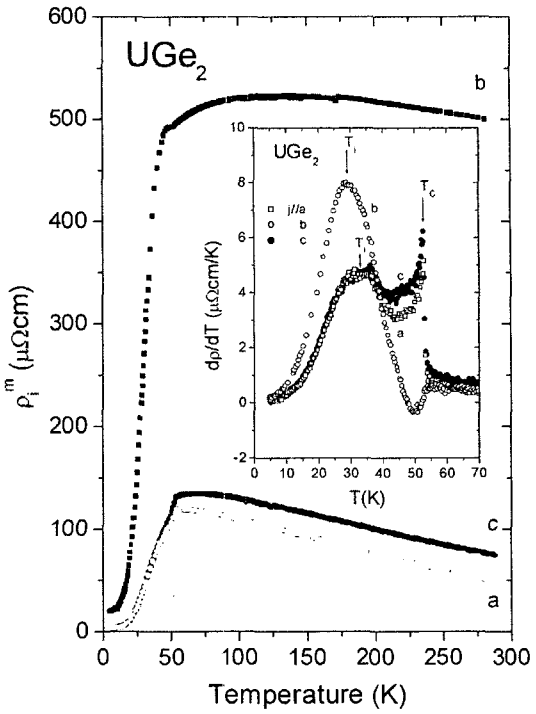

Fig. 5. Only the magnetic part of the resistivity ρ_i^m, where i = a, b and c, as a function of temperature measured for sample I. The inset shows the temperature derivative of the resistivity, $d\rho(T)_i/dT$, determined for sample II.

[7] which had a similar fashion with a strong curvature of the σ^2 versus B/σ function for $B/\sigma \rightarrow 0$. In both situations a strong curvature of this function close to the transition temperature does not allow to determine precisely the Curie point by this method.

2.3. Electrical resistivity and magnetoresistivity

Electrical resistivity was measured on two oriented single crystals I and II, cut off from different batches. We have also measured $\rho(T)$ for the polycrystalline sample of $ThGe_2$, considering it as a diamagnetic reference for UGe_2. The measured $\rho(T)$ curve between 4.2 and 300 K for this compound is typically of metallic character, i.e. it shows a "hockey stick" shape. Fig. 5 demonstrates the extracted magnetic part of the UGe_2 resistivity ρ_i^m (i = a, b, c) versus T, obtained for sample I by subtraction from the total $\rho_i^t(T)$ the phonon part by using in this purpose the only temperature dependence of the reference resistivity of $ThGe_2$.. From this figure it is clear that along the hard directions, where j//b and j//c, the slope $d\rho_i^m/dT$ becomes negative, which reminiscent a Kondo effect, while for the j//b configuration this is not so apparent. It appears that the $\rho_b(T)$ curve above T_C goes through a wide maximum at 175 K. In this figure the temperature derivative of the resistivity found for three main axes is

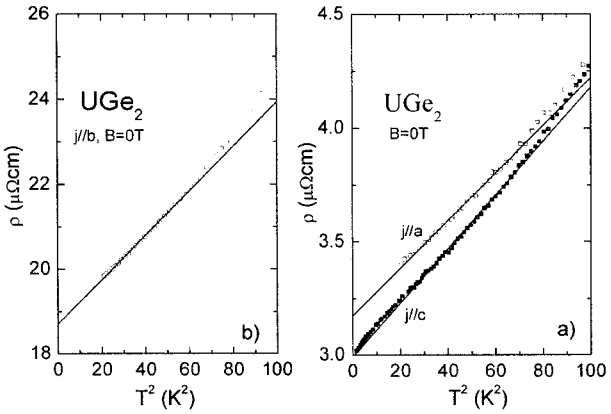

Fig. 6. The ρ_i vs T^2 Fermi-liquid function determined for three main crystallographic directions.

also displayed in the inset. If T_C is exactly same for all three directions, the inflection point T_i determined by a maximum in $d\rho(T)/dT$ for the b-axis is found at lower temperature than that for the a and c-axes. It has been discussed in Ref. 4 that T_i corresponds to the characteristic temperature T^*.

As shown in Fig. 6, the electrical resistivity measured along the three main crystallographic directions can be fitted below about 8 K to the expression: $\rho_i(T) = \rho_{i0}(T) + A_i T^2$ (i = a, b, c), following of which may indicate the presence of both the magnetic fluctuations (Fermi-liquid state) and/or spin wave excitations. The latter due to the Ising type of the magnetization [12] somehow precludes the latter contribution. The obtained parameters are presented in Table 2. Previously the values of the coefficient A_i were given only for ρ_a (j||a) [2], for ρ_{poly} [13] and for ρ_x (not defined direction) [14, 15]. All there reported data are close to those given in Table 2 for the a- and c-axes. As this Table indicates, the A_b value is almost 5 times larger than those of A_a and A_c. This kind anisotropy in the A_i coefficients should be explained in the future studies. As is known, the magnitude of the coefficient A is connected with the strength of magnetic fluctuations and with a magnitude of T_{sf} ($A \sim T_{sf}^{-2}$). For further discussion see also Ref. 4.

In Fig. 7 we have plotted the magnetoresistivity $\Delta\rho/\rho_0$, defined as the ratio $[\rho(T,B)-\rho(T,0)]/\rho(T,0)]$, against

Table 2. Fermi-liquid characteristics of a UGe_2 single crystal (sample I)

ρ_i	ρ_0 (μΩcm)	A_i (μΩcm/K²)
a	3.2	0.011
b	18.7	0.052
c	3.03	0.012

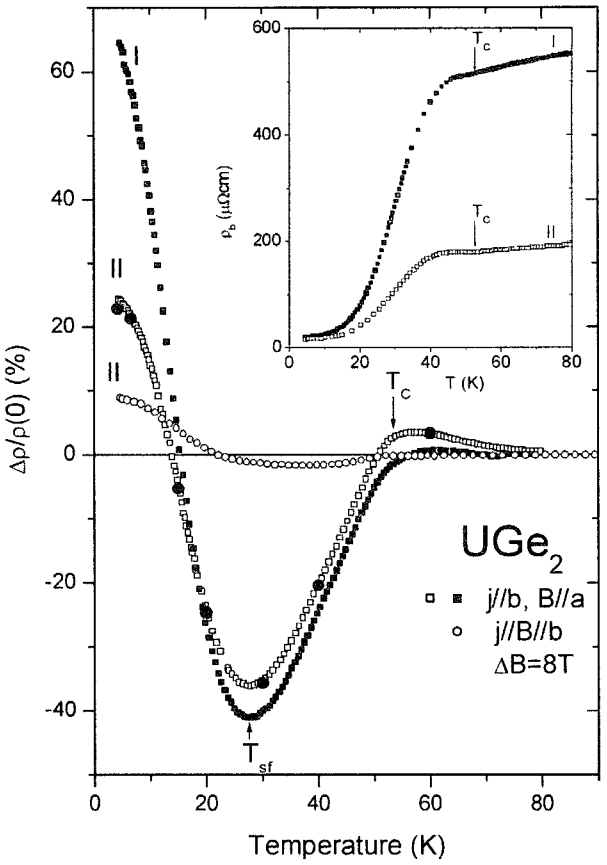

Fig. 7. Transverse (for samples I and II, solid and open squares, respectively) and Longitudinal (sample II, open circles) magnetoresistivity, $\Delta\rho/\rho(0)$ vs. T, measured for the current j flowing along the b-axis and at 8 T applied perpendicular to the current direction. The large solid circles are the data taken from the $\Delta\rho/\rho(0)$ vs B curves at 8 T for sample II (not shown here). The inset shows the different temperature behaviour of ρ_b for samples I and II.

temperature for both single crystals measured. Practically the difference between samples I and II exists only at low temperatures. This results together with those earlier published in Ref. 5 for sample I clearly indicate the presence of huge effect in $D\rho/\rho_0(T)$ at T_{sf} near the characteristic temperature T^* of UGe_2. In the inset to this figure the $\rho_b(T)$ curves measured for samples I and II are presented. Although there were only negligible differences in the lattice parameters between the samples [see for the values in Ref. 5], the electrical resistivity measured for the current j//b showed ρ_b (300 K) almost to be three times larger for sample I compared to that for sample II, but with almost the same value of $\rho_0 \approx 20$ mWcm for both the samples. Despite the higher value of RRR for the former sample (31.5 against 14.0) a quite good agreement with the $\rho(T)$ data of Onuki *et al.* [8] was just found for sample II with lower RRR value. It should

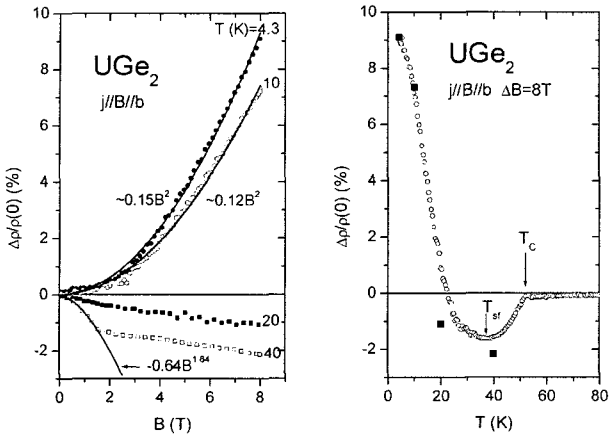

Fig. 8. The longitudinal magnetoresistivity (LMR) measured at various temperatures as a function of applied field B up to 8 T (left hand panel) and measured at 8 T as a function of temperature T up to 80 K (right hand panel).

be mentioned that the RRR values of the single crystals reported in Ref. 8 are about an order of magnitude higher than those presented here indicating a very good quality of samples used. Nevertheless, the character of ρ(T) and the Fermi -liquid parameters are practically the same for both measurements. In accordance to the data of Onuki *et al.* [8] there is well seen a hump in this dependences just below T_C, as marked by the arrows in the inset of Fig. 7. The origin of the hump, observed only for the j//b configuration, is still not explained. This would be so far the only eventual signal of the existence of a spin density wave (SDW) behaviour in UGe_2, but detected only along the expanded b axis. The latter has been predicted theoretically [16] to occur at temperatures below T^*, but still not being confirmed experimentally by neutron scattering methods [1, 17].

Finally in Fig. 8 there is demonstrated for the first time the field (figure a) and temperature (figure b) dependencies of $\Delta\rho/\rho(0)$, measured for sample II but in the configuration j//b//B, yielding the longitudinal magnetoresistivity (LMR) along the magnetization hard direction b. It becomes clear in Fig. 6 that the effect is considerably smaller in comparison to that found in TMR, especially at the temperature range where the MR becomes negative. As is seen in Fig. 7a, the field dependences of at temperatures 20 and 40 K are almost linear in B, while at lower temperatures it changes as a B^2. Interestingly, as shown in figure (b), the temperature dependence of $\Delta\rho/\rho(0)$ exhibits a similar character as that of TMR, but the LMR value at the negative maximum, occurring also near T^*, is only 5% of that observed at T_{sf} of TMR. For LMR, the latter temperature seems to be shifted a little to higher temperatures, compared to that of TMR. Again, no effect in LMR is seen at T_C. The difference in LMR and TMR is shown in Fig. 6. It probably arises from a large anisotropy existing in the magnetic fluctuations depending on the mutual configuration of the current and field, being further in the relation to the crystallographic axes.

3. Discussion

The properties which have been revealed under pressure in UGe_2, where the relatively strong and highly anisotropic FM allows or may even induce SC, can not be understand in terms of existing theories and need a novel concept [18]. A new mechanism of simultaneous appearance of ferromagnetism and superconductivity based on interaction of electrons mediated by localized moments has recently been proposed by Suhl [19] and Abricosov [20]. In their model the simultaneous onset of magnetic order and superconductivity may be explained as a coupling of two conduction electrons *via* one localized moment. Here, especially on the basis of recently performed studies aimed at the determination of the Fermi surface (FS) in the paramagnetic state of UGe_2 [21], it becomes clear that FM comes from the fairly well localized $5f^{3-n}$ electrons, with $n \approx 1$, while Cooper pairs probably originate from medium heavy quasiparticles (the electronic heat capacity coefficient $\gamma(0) = 30$ mJ/K^{2}mol [22]) formed by the hybridized $5f^n$ electrons with the conduction electrons, which microscopically emphasizes the double role of the 5f electrons in this compound. In the circumstances of existing a large internal field due to FM, the only so called nonunitary triplet state, being free from the Pauli limit, can be taken into account as the SC-state [23]. Thus spin (magnetic) fluctuations as a mediated pairing mechanism in the case of such a state has been proposed which seems to be in line with the discovery in Ref. 5 of the huge magnetoresistivity effect detected deeply in the ferromagnetic state and being confirmed here by carrying out the MR measurements on the different single-crystalline samples of UGe_2. The decrease of the resistivity by about 40% at the temperature close to T^* could only be possible by depressing magnetic fluctuations by an applying a field of 8 T along the easy axis a with the current flowing along the most hard and largely elongated b-axis. Furthermore, the more detailed studied under pressure of the coefficient A_b in the Fermi-liquid formula just along the b-axis in the vicinity of p_C^* can probably give more arguments about the role of the magnetic fluctuations, detected along the b-axis, in the creation of SC in UGe_2. As we have signaled above this coefficient at ambient pressure is about 5 times larger than the remaining A_a and A_c coefficients. If only a

modulation of FM by SDW will be detected in the future, one then can think that the fluctuations of coupled charge density- (CDW) and spin density-(SDW) waves, postulated in the literature [16], are just those which are responsible for the MR effect, described in this work. Very recently a microscopic model for ferromagnetism of UGe_2, which takes into account the competition of the orbital degeneracy of the 5f electrons as well as the anisotropies of the CEF and hopping terms has been proposed [24]. In consequence one of possible magnetic moment arrangements is the transverse to the (ac)-plane a SDW component but with a small magnetic amplitude of 0.01 μ_B, the value considerably lower than the resolution of neutron scattering experiment [17], but in accordance with results of positive muon spin relaxation measurements performed on a UGe_2 single crystal [25]. In the latter work in addition to the well-known localized 5f-electron density responsible for the ferromagnetic bulk properties, the existence of itinerant quasistatic magnetic correlation with magnetic moment of about 0.02 μ_B has been found. All these findings seems to be supported by the recently observed electronic-kind transition near T^* in the Hall effect experiment performed on a single crystal of UGe_2 [26].

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