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Pressure-induced antiferromagnetism in UPt

K. Prokeš*
Hahn-Meitner-Institute, SF-2, Glienickerstrasse 100, Berlin, Germany

N. V. Mushnikov† and T. Goto
Institute of Solid State Physics, University of Tokyo, Kashiwanoha, Kashiwa-shi, Chiba-ken, Japan

T. Tahara, M. Ito, T. Goko, F. Nakamura, and T. Fujita
Department of Quantum Matter, ADSM, Hiroshima University, Higashi-Hiroshima, Japan

F. Honda and G. Oomi
Department of Mechanical Engineering and Materials Science, Kumamoto University, Kumamoto, Japan

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UPt orders ferromagnetically at ambient pressure at 28 K. Upon increasing pressure, an additional magnetic phase transition appears around 17 K. While the upper transition behaves with field as a typical paramagnetic-ferromagnetic transition, the lower one exhibits antiferromagnetic behavior. With pressure both transitions shift towards lower temperatures. Around 1.5 GPa the upper magnetic transition completely disappears and only the lower persists up to ~4 GPa. The electrical resistivity measured up to 8.0 GPa shows that structural transformation occurs in UPt under pressure.

I. INTRODUCTION

Ferromagnetic UPt has attracted considerable attention over the last three decades[1] due to unusual magnetic properties under pressure. A strong depreciation of the magnetization with pressure and pressure-invariant magnetic phase transition temperature of about 27 K has been reported for UPt.[2–4] The ambient-pressure saturation magnetization which amounts at 4.2 K to about 0.45 μ_B/U was reported to be strongly sample dependent[4] and in clear contrast with the neutron-diffraction results of Frings *et al.*[5] of $1.1 \pm 0.4\,\mu_B$/U. Recently, magnetic measurements on small single crystals led to a moment of 1.01 μ_B/U.[6]

UPt is formed incongruently by a solid-state reaction between U and UPt_2 (Ref. 2) and it is reported to form in a monoclinic structure similar to UIr.[7,8] Almost all publications dealing with UPt mention two magnetic phase-transition temperatures to ferromagnetic order at 19 and 27 K.[2,3] Neutron-diffraction work by Frings *et al.*[5] and by Franse *et al.*[9] suggested that each of the two magnetic transitions is connected with a different structural phase. The upper magnetic phase transition is connected with PdBi-type ordered UPt and the lower one with ordering in some distorted structure. Moreover, the latter structure was reported to be transformed under pressure to the former one. This resulted in a decrease of the magnetic moment.[3,5,9] Magnetization measurements under hydrostatic pressure up to 0.77 GPa and in fields up to 8 T by Frings *et al.*[4] revealed that a metamagnetic transition evolves under pressure at low temperatures. However, these authors later concluded on the basis of low-temperature neutron-diffraction experiments under pressure up to 0.45 GPa that no evidence for pressure-induced antiferromagnetic phase was found.[5]

Rather peculiar behavior under pressure has motivated us to reinvestigate magnetic and transport properties of this compound under pressure. Here we report on magnetic measurements up to 1.3 GPa between 1.6 and 200 K and on the electrical resistivity up to 8.0 GPa between 4.2 and 300 K of a polycrystalline UPt.

II. EXPERIMENT

Polycrystalline UPt was prepared by evaporating In from U-Pt-In melt containing the stoichiometric amount of U and Pt. The quality of the resulting material has been inspected by electron microprobe analysis and by powder x-ray diffraction. The material was found to consist of stoichiometric UPt and traces (<3%) of the second phase which was found to be U rich, most probably UO_2, UO or U. Rietveld-type analysis of the x-ray-diffraction pattern suggests that UPt crystallizes in the monoclinic structure with the space group $P2_1$ (No. 4, $Z=8$). No other structural phases of UPt were identified. Structural parameters, which agree well with literature values,[5,8] and further details concerning the sample preparation can be found in Ref. 10.

For magnetic measurements, the sample (cylinder with a diameter of 2.9 mm and a length of 3.4 mm) has been cut by spark erosion. It has been placed in a Teflon capsule filled with a pressure-transmitting medium (1:1 mixture of FC 70 and FC 77 Fluorinert) and compressed in a small Cu-Ti clamp-type cell. Due to compensating contributions of Ti and Cu, this alloy has very low magnetic susceptibility and is suitable for magnetic studies. Nevertheless, all the data have been corrected for the background magnetization. Pressure was calibrated by measuring the Meissner effect of Pb. A conventional ^{4}He insert and a superconducting magnet have been used and the signal detected by the extraction method. Further details on the experimental apparatus and data processing together with error analysis can be found in Ref. 11.

The electrical resistivity was measured on small bar-

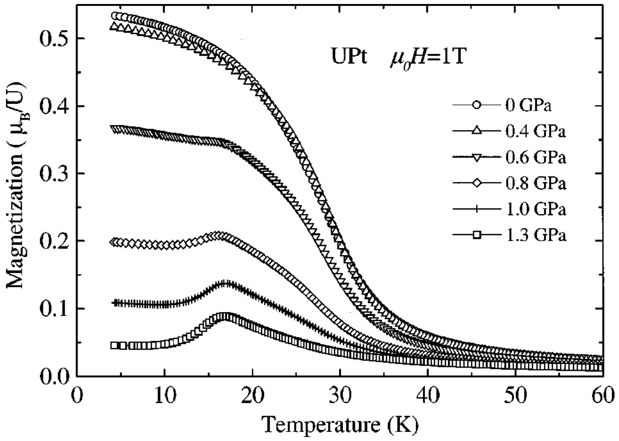

FIG. 1. Temperature dependences of magnetization measured at various pressures on bulk sample of UPt in field of 1 T.

shaped samples by a standard dc four-point method in the 4.2–290 K temperature range. The electrical contacts were established by gold wires fixed to the surface by a silver paint. To produce a pressure, two apparatus were employed. The first, installed at the Hiroshima University uses a cube-anvil device with a maximum pressure of 8.0 GPa and the second installed at the Kumamoto University a standard piston-cylinder device made of Cu-Be alloy with a maximum pressure of 2.2 GPa.[12] In the latter case, the load is kept constant and regulated automatically with a deviation of less than 1% so that there is no significant pressure difference on cooling and heating runs.

III. RESULTS

A. Magnetic measurements

In Fig. 1, temperature dependences of the magnetization measured in a field of 1 T at various pressures up to 1.3 GPa are shown. It is quite clear that UPt orders in this field at ambient pressure magnetically below 28.6 K (determined by a maximum slope in $\partial M/\partial T$). The transition temperature increases with the application of field suggesting that ferromagnetism exists in UPt below $T_C = 28.6\,\mathrm{K}$. If measured in low fields (below 0.1 T), our sample exhibits also a weak additional magnetic anomaly around 21 K which was sometimes reported in the literature to be the dominating phase transition.[1,2,13] As the applied pressure increases, the magnetization below T_C is reduced and a new anomaly around 17 K appears. At the highest pressure applied, at 1.3 GPa nearly no sign of the upper transition (we denote it as $T_H = T_C$) is left and the shape of the anomaly at the lower temperature strongly resembles that of an antiferromagnetic (AF) phase transition (we denote this temperature as T_L). Both phase transitions shift linearly with pressure P towards lower temperatures with a rate of $\partial T_H/\partial P = -2.0 \pm 0.3\,\mathrm{K/GPa}$ and $\partial T_L/\partial P = -0.8 \pm 0.3\,\mathrm{K/GPa}$. The latter value suggests that UPt should become paramagnetic at ~20 GPa.

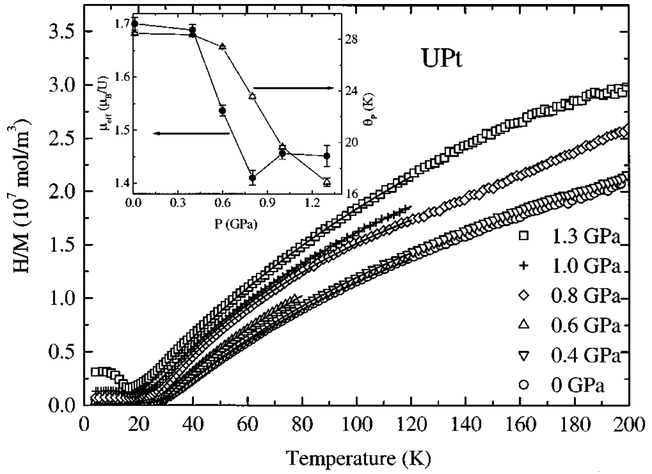

FIG. 2. Temperature dependences of the inverse magnetic susceptibility ($\chi = M/H$) measured on bulk piece of UPt in 1 T at various pressures. The solid lines represent best fits to a modified Curie-Weiss (MCW) law. In the inset results of the fits are shown.

In Fig. 2 we show the temperature dependences of the inverse magnetic susceptibility ($\chi = M/H$) measured in field of 1 T at several pressures. It is evident that application of pressure decreases the effective magnetic moment μ_{eff} and also reduces paramagnetic Curie temperature θ_P. Both are reduced with applied pressure in a nonlinear way, which reflects the appearance of the new pressure-induced AF phase. The pressure dependences of the effective moment μ_{eff} and paramagnetic Curie temperature θ_P determined from modified Curie-Weiss fits performed, except for $P = 0.6\,\mathrm{GPa}$, in the temperature region between 40 and 120 K are shown in the inset of Fig. 2. It is clear that between 0.4 and 0.8 GPa magnetic properties of UPt undergo a drastic change. For instance, at 1.3 GPa, μ_{eff} drops by 15% and θ_P by 40% of its ambient-pressure value. The temperature-independent term χ_0 shows a drastic change in this pressure region as well (not shown).

The field dependence of the magnetization measured at various temperatures at 1.3 GPa is presented in Fig. 3. For the sake of clarity, data have been shifted for each subsequent lower temperature by $0.05\,\mu_B/\mathrm{U}$ starting from 22 K. Clearly, below 16 K a metamagneticlike transition is present around the critical field $B_C = 2.6\,\mathrm{T}$. Temperature dependence

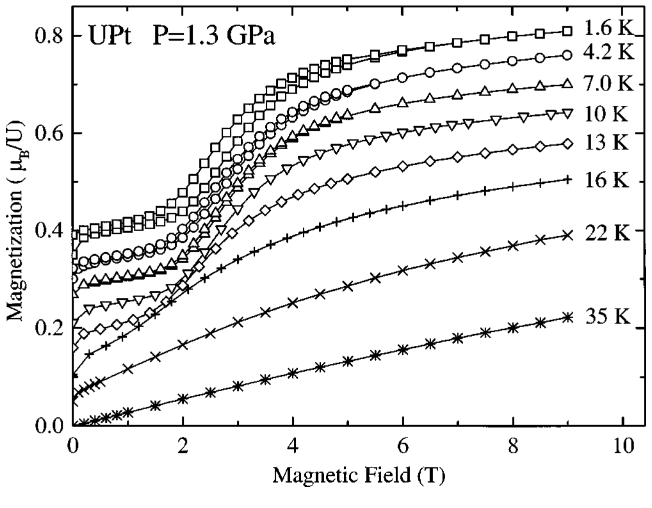

FIG. 3. Field dependences of the magnetization of UPt measured at 1.3 GPa at various temperatures in fields up to 9 T. For the sake of clarity, data have been shifted for each subsequent temperature by $0.05\mu_B/U$ starting from 22 K (curve at 35 K not shifted).

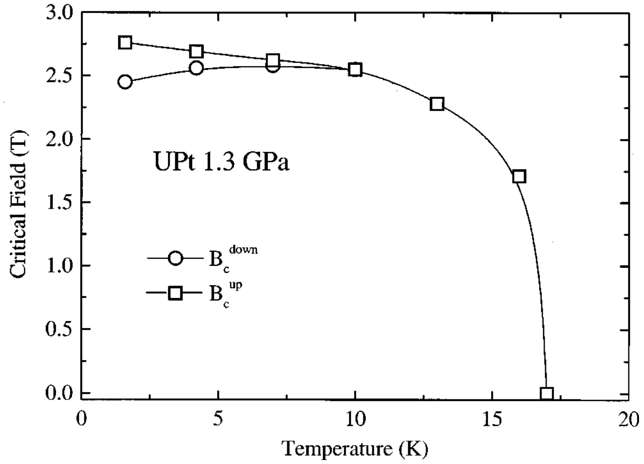

FIG. 4. Temperature dependence of the critical field of the metamagnetic transition marking the transition from the AF to the F state of UPt at 1.3 GPa (deduced from data shown in Fig. 3).

of the critical field of this transition, which exhibits below 10 K a hysteretic behavior, is shown in Fig. 4. The position of the midpoint of B_C (average of up and down branches) decreases with increasing temperature and becomes zero around 16 K.

In Fig. 5, the field dependence of magnetization measured at 4.2 K at various pressures up to 1.3 GPa and in fields up to 9 T is shown. Upon application of pressure, the original magnetization curve, which has at ambient pressure shape resembling strongly ferromagnetic material, starts to change its shape for $P \sim 0.4\,\mathrm{GPa}$. Above this pressure, a metamagneticlike transition evolves around $B_C = 2.6\,\mathrm{T}$. There is a small hysteresis with field sweeping up and down through B_C. The ferromagnetic component below the metamagnetic transition decreases very strongly with pressure. At the same time, the magnetization step across the metamagnetic transition increases. The magnetization attained at 9 T, $\mu_{9\mathrm{T}}$, decreases nonlinearly with increasing pressure. At pressures below 0.6 GPa it decreases with a rate of $\partial\mu_{9\mathrm{T}}/\partial P_{<0.6\mathrm{GPa}} = -0.028\,\mu_B/\mathrm{U\,GPa}^{-1}$. At higher pressures, above 0.8 GPa, it decreases faster with a rate of $\partial\mu_{9\mathrm{T}}/\partial P_{>0.8\mathrm{GPa}} = -0.094\,\mu_B/\mathrm{U\,GPa}^{-1}$. The magnetization value linearly extrapolated to zero field, $\mu_{0\mathrm{T}}$, also decreases with pressure in similar way with rates $\partial\mu_{0\mathrm{T}}/\partial P_{<0.6\mathrm{GPa}} = -0.04\,\mu_B/\mathrm{U\,GPa}^{-1}$ and $\partial\mu_{0\mathrm{T}}/\partial P_{>0.8\mathrm{GPa}} = -0.11\,\mu_B/\mathrm{U\,GPa}^{-1}$. Extrapolation of the latter tendency towards higher pressures leads to the conclusion that magnetic moments should be lost around 4 GPa. Above this pressure, UPt is expected to be paramagnetic down to lowest temperatures. All these dependences are depicted in Fig. 6. The metamagnetic-transition field B_C at 4.2 K, gets reduced with pressure by a rate of -0.2 T/GPa.

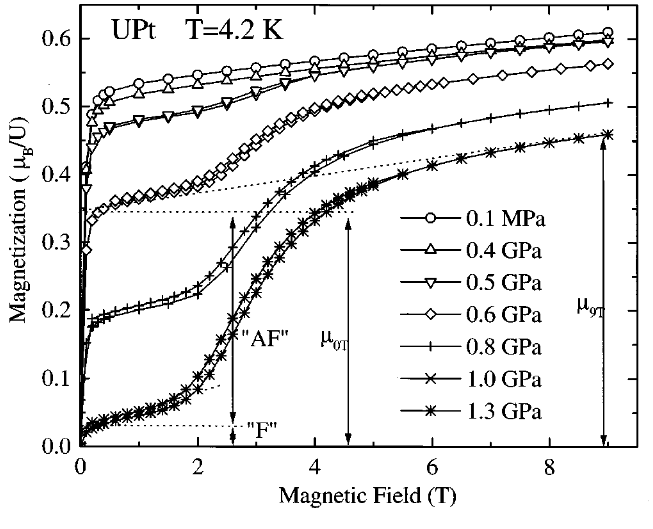

FIG. 5. Field dependences of the magnetization of bulk UPt measured at 4.2 K and at various pressures in fields up to 9 T.

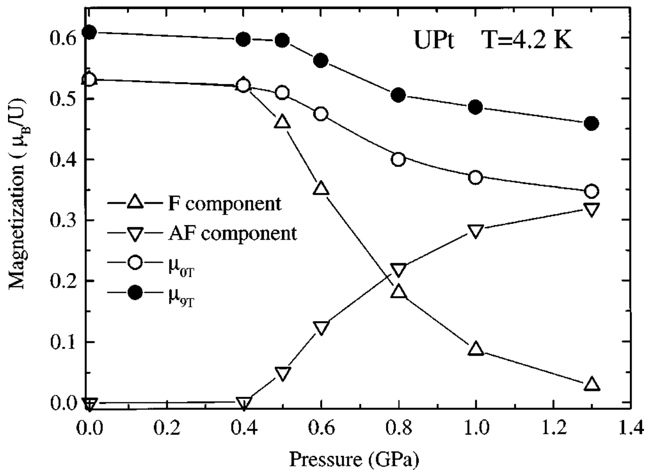

FIG. 6. Pressure dependence of the magnetization of UPt at 4.2 K in pressures up to 1.3 GPa deduced from data shown in Fig. 4. Legend: AF is the magnetization gain due to metamagnetic transition, F is the ferromagnetic component, $\mu_{0\mathrm{T}}$ is the magnetization extrapolated from high fields to zero field, $\mu_{9\mathrm{T}}$ is the magnetization at 9 T.

In Fig. 7 we show the field dependence of the magnetization measured at a few pressures at 22 and 35 K. Clearly, no metamagnetic transition is present in either data sets. However, suppression of the magnetization values with pressure is apparent. At 1.3 GPa, however, a very small ferromagnetic intercept can be still discerned for data taken at 22 K. This is absent in data taken at 35 K for all pressures in agreement with magnetic susceptibility data.

B. Electrical resistivity

In Fig. 8 the low-temperature detail of the temperature dependence of the electrical resistivity measured under pressure up to 8 GPa is shown. In Fig. 9 we show the whole temperature region measured. At ambient pressure we observe the resistivity curve that is very similar to literature results.[4,10,14] At high temperatures, the electrical resistivity slightly increases with lowering temperature which is usually interpreted as to be due to hybridization of conduction electrons with $5f$ sates. Around 150 K it exhibits a maximum and starts to decrease. Below approximately 28 K the electrical resistivity drops drastically due to the appearance of magnetic order. This characteristic albeit with slightly modified high-temperature slope and alternated position of the maximum, is observed for all measurements performed under pressure $P \leqslant 0.6\,\mathrm{GPa}$. At higher pressures but lower than 1.4 GPa there are two transitions visible as change in the

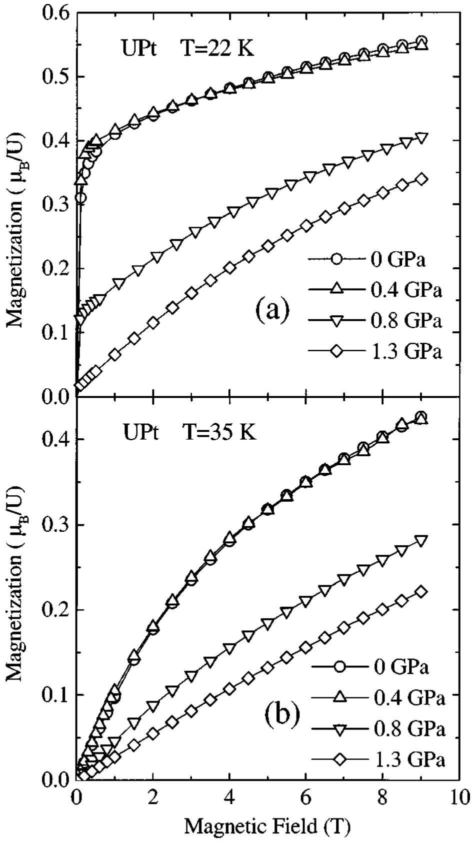

FIG. 7. Field dependence of the magnetization of bulk UPt measured at 22 K (a) and at 35 K (b) at various pressures in fields up to 9 T.

$\rho(T)$ slope. One is observed at progressively lower temperatures with respect to the ambient-pressure value as the pressure increases. The other transition is seen at about 17 K. Between 1.4 and 3.0 GPa, only the lower transition that shifts with pressure towards lower temperature is seen on $\rho(T)$. Above 3.0 GPa the resistivity monotonically decreases with temperature in the whole temperature range without any sign of a magnetic phase transition (this is in agreement with magnetic measurements suggesting loss of magnetic order around 4 GPa). The magnetic phase transitions determined from resistivity curves show tendencies identical to results obtained from magnetic measurements. Moreover, in resistivity curves taken at 0.6, 1.0, 1.4, and 1.5 GPa one can observe hysteresis between cooling and warming branches with the middle point at $\sim$75, $\sim$180, $\sim$280, and at $T > 300$ K, respectively. Consequently, the room-temperature electrical resistivity shows a significant increase between 1.0 and 1.4 GPa. Such a behavior could be interpreted in terms of a structural phase transformation.

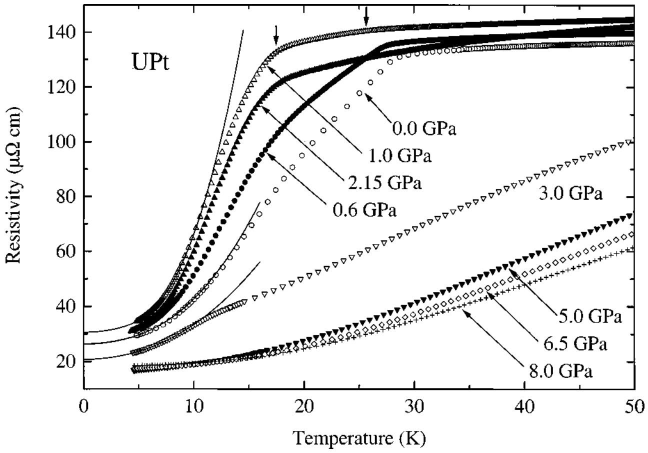

FIG. 8. The low-temperature detail of the temperature dependence of the electrical resistivity of UPt uder pressure up to 8 GPa. Arrows at the top idicate the two magnetic phase transitions seen for pressures $0.6 < P < 1.4$ GPa. Solid lines through symbol taken at ambient pressure, 1.0 and 3.0 GPa are the best fits to expression given in the text.

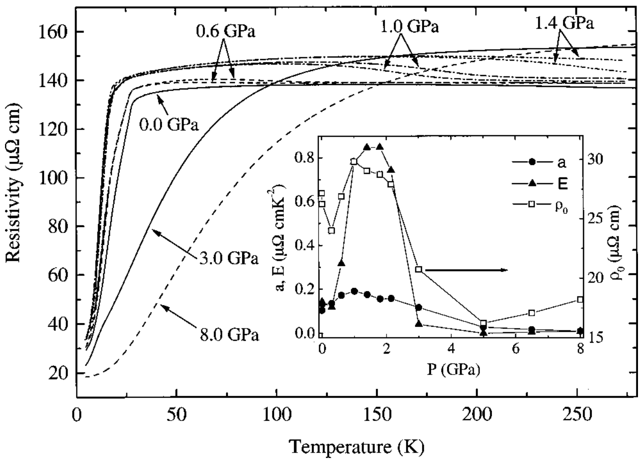

FIG. 9. The temperature dependence of the electrical resistivity of UPt uder pressure up to 8 GPa. In the inset we show graphically results of the fits to expression given in the text keeping $\Delta = 25$ K (three fits shown in Fig. 8).

The low-temperature parts of UPt resistivity can be well fitted by expression $\rho = \rho_0 + aT^2 + E\Delta T(1 + 2T/\Delta)\exp(-\Delta/T)$, where E depends on the spin-disorder resistivity and the electron-magnon coupling constant and Δ denotes the energy gap between the upper boundary of majority spin subband and the Fermi energy. Let us note that we have used successfully this expression for a description of the specific heat[10] and also for the saturation magnetization of small single crystals.[6] The same temperature dependence is encountered for all measurements and the best fit gives $\Delta = 20-30$ K for all pressures. Results of the best fits to the aforementioned expression in the low-temperature region below 9 K, ρ_0, a, and E, keeping $\Delta = 25$ K are shown graphically in the inset of Fig. 9. All fitting parameters show a maximum (ρ_0 at $\sim$1.0 GPa, a at $\sim$1.0 GPa, and E at $\sim$1.6 GPa, respectively) before declining at higher pressures. In the case of ρ_0 a small increase is observed above 5.0 GPa.

IV. DISCUSSION AND CONCLUSION

It is well known that in $5f$ intermetallics with the shortest U-U separation falling into the critical Hill's region,[15] $5f$ electron states form more or less narrow, strongly correlated bands pinned at or near the Fermi level E_F. This is reflected in UPt, for instance in the enhanced low-temperature specific-heat coefficient γ, which amounts to 105 mJ/mol K^2,[4,9,10,13] temperature-independent high-temperature resistivity,[4,10,14] and in enhanced temperature-independent parameter χ_0 reflecting a high density of states at the Fermi level. As U atoms are brought closer, the originally narrow bands get wider. Values of γ and μ_{eff} are usually reduced and the temperature dependence of the electrical resistivity at high temperatures behaves more in a metallic way (i.e., it increases with increasing temperature). Of course, all polycrystalline data of UPt are strongly modified with respect to data obtained on single crystals due to mixing of different Curie-Weiss (CW) branches because of strong magnetic anisotropy.[5,6,10] Therefore, it is in general difficult to compare polycrystalline data taken on different samples (due to texture). Here we compare, however, magnetic data taken on one sample without remounting it and obtained by fitting under the same conditions. Therefore, we believe that we can draw conclusions about the influence of pressure on the magnetic properties of UPt.

Reduction of μ_{eff} in UPt with pressure is understandable quite easily because its application reduces interatomic distances and consequently, increases hybridization between $5f$ electron states with other states in the structure. This, in turn, delocalizes U moments. However, one would expect a smooth decrease of μ_{eff} with pressure in contrast to observed step between 0.4 and 0.8 GPa. A similar steep decrease is seen also in the pressure dependence of θ_P. Moreover, χ_0 shows also a drastic change in this pressure region. These results together with low-temperature magnetization data (occurrence of a metamagneticlike transition at $B_C = 2.6\,\mathrm{T}$) strongly suggest that a magnetic phase transformation occurs in UPt under pressure. The original low-pressure phase which is stable for $P < 0.4\,\mathrm{GPa}$ is ferromagnetic (F) with $T_C = 28.6\,\mathrm{K}$ at 1 T and at ambient pressure. The high-pressure phase, stable for $1.5\,\mathrm{GPa} < P < \sim 4$ GPa is antiferromagnetic (AF) with $T_N \sim 16\,\mathrm{K}$ at 1 T and at 1.5 GPa. Above ~4 GPa UPt does not order magnetically and between 0.4 and 1.5 GPa it consists of an admixture of both the aforementioned phases. The ratio between both phases changes with pressure in favor of the AF phase. The content of the low-pressure phase decreases by 17% per 0.1 GPa between 0.6 and 1.0 GPa. Further, we can conclude that the saturation moment $\mu_{\mathrm{sat}}^{\mathrm{F}}$ of the F phase is higher than that of the AF phase $\mu_{\mathrm{sat}}^{\mathrm{AF}}$. Knowing that the anisotropy of UPt is of an uniaxial type[5,6,10] these values are estimated to be $\mu_{\mathrm{sat}}^{\mathrm{F}} = 1.06\,\mu_B/\mathrm{U}$ and $\mu_{\mathrm{sat}}^{\mathrm{AF}} = 0.70\,\mu_B/\mathrm{U}$ (for the AF phase we supposed for simplicity no pressure dependence). The former value agrees well with single-crystal data[6] and neutron-diffraction results.[5,9]

Both magnetic phase transition temperatures decrease with pressure as indicated by magnetic measurements ($\partial T_H/\partial P = \partial T_C/\partial P = -2.0 \pm 0.3\,\mathrm{K/GPa}$ and $\partial T_L/\partial P = \partial T_N/\partial P = -0.8 \pm 0.3\,\mathrm{K/GPa}$). The latter value suggests that UPt should become paramagnetic at ~20 GPa, i.e., at a pressure that is much higher than that indicated by the extrapolation of the magnetic moment to higher pressures and that indicated by electrical resistivity (in both latter cases it amounts to ~4.0 GPa). There are two main possibilities to explain this disagreement. In our analysis we assumed a linear decrease of T_N with pressure in the whole pressure range. Such an approach might not be applicable because it neglects the pressure dependence of the magnetic moment magnitudes which in turn modifies the strength of magnetic interactions. It also neglects the fact that above ~1.5 GPa the phase transformation is finished and above this pressure we deal only with one phase. If for whatever reasons the compressibility of the low-pressure phase is higher than that of the high-pressure phase, the former phase would provide a spacer, eliminating partly the effect of increased pressure on the latter phase. In this case one should expect above 1.5 GPa a different, presumably much higher, value of $\partial T_N/\partial P$. This is indeed suggested by electrical resistivity results leading to $\partial T_N/\partial P \cong -4.3 \pm 2.0\,\mathrm{K/GPa}$. Most probably, a combination of all aforementioned mechanisms leads to a collapse of the magnetic order in UPt with pressure.

As was noticed in previous neutron-diffraction experiments,[5,9] new Bragg reflections that are not consistent with the original crystal structure appearing in the neutron-diffraction pattern at low temperatures upon application of a pressure of 0.7 GPa. Although some of these reflections or at least a certain portion of intensity of these reflections can be attributed to magnetic order (it is not clear whether AF or F) some of them are present under pressure even at room temperature.[3] However, after removing the pressure and annealing at 300 K they disappear. These results were interpreted in terms of a structural phase transformation that occurs in UPt under pressure. Our magnetic and electrical resistivity measurements strongly support this idea. First, the high-temperature part of the magnetic susceptibility undergoes a drastic change (according to fitted parameters) above 0.4 GPa. Second, above the same pressure also the low-temperature dependence of magnetization changes entirely its character. While at low pressures it shows typical ferromagnetic behavior at higher pressures this behavior is changed to an antiferromagnetic one. Third, this is corroborated by the appearance of the metamagneticlike transition. The pressure development of the magnetic phase transition temperatures and of the magnetization curves suggests that there is a gradual change in a volume ratio between F and AF phases with pressure. All these results suggest that a magnetic phase transformation that occurs in UPt is intimately connected to a structural transformation. Perhaps the strongest indication for a change in the crystal structure of UPt can be found in the electrical resistivity. In curves taken at 0.6, 1.0, 1.4, and 1.5 GPa one sees clear hysteresis at progressively increasing temperatures suggesting that the applied pressure extends the stability of the pressure-induced structural phase to higher temperatures. This finding is in agreement with the neutron results showing[5,9] reversible temperature changes of some Bragg reflections under pressure.

In conclusion, we report on the magnetic properties of UPt polycrystalline sample under pressure up to 1.3 GPa and on the electrical resistivity up to 8.0 GPa. We found clear evidence for the pressure-induced ferromagnetic–antiferromagnetic transition which is due to structural transformation.

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[*] On leave from Department of Electron Structures, Charles University, 12116 Prague 2, The Czech Republic.

[†] Permanent address: Institute of Metal Physics, S. Kovalevskaya 18, 620219 Ekaterinburg, Russia.

[1] B. T. Matthias, C. W. Chu, E. Corenzwit, and D. Wohlleben, Proc. Natl. Acad. Sci. U.S.A. **64**, 459 (1969).

[2] J. G. Huber, M. B. Maple, and D. Wohlleben, J. Magn. Magn. Mater. **1**, 58 (1975).

[3] P. H. Frings, J. J. M. Franse, F. R. de Boer, and A. A. Menovsky, J. Magn. Magn. Mater. **31–34**, 240 (1983).

[4] P. H. Frings and J. J. M. Franse, J. Magn. Magn. Mater. **51**, 141 (1985).

[5] P. H. Frings, C. Vettier, A. Dommann, F. Hulliger, and A. A. Menovsky, Physica B **156&157**, 832 (1989).

[6] K. Prokeš, T. Fujita, E. Brück, F. R. de Boer, and A. A. Menovsky, Phys. Rev. B **60**, R730 (1999).

[7] A. Dommann, F. Hulliger, T. Sigrist, and P. Fisher, J. Magn. Magn. Mater. **67**, 323 (1987).

[8] A. Dommann and F. Hulliger, Solid State Commun. **65**, 1093 (1988).

[9] J. J. M. Franse, A. A. Menovsky, A. de Visser, P. H. Frings, and C. Vettier, J. Magn. Magn. Mater. **70**, 351 (1987).

[10] K. Prokeš, J. C. P. Klaasse, I. H. Hagmusa, A. A. Menovsky, E. Brück, F. R. de Boer, and T. Fujita, J. Phys.: Condens. Matter **10**, 10 643 (1998).

[11] K. Koyama, S. Hane, K. Kamishima, and T. Goto, Rev. Sci. Instrum. **69**, 3009 (1998).

[12] G. Oomi and T. Kagayama, Physica B **239**, 191 (1997).

[13] C. A. Luengo, M. B. Maple, and J. G. Huber, J. Magn. Magn. Mater. **3**, 305 (1976).

[14] J. Rodriguez Fernandez, M. A. Argüelles, J. I. Espeso, J. C. Gomez Sal, and K. A. McEwen, Physica B **86–88**, 87 (1997).

[15] H. H. Hill, in *Plutonium and Other Actinides*, edited by W. N. Miner (AIME, New York, 1970).