Magnetic irreversibility in the antiferromagnetic state of UPt$_2$Si$_2$

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We present a detailed investigation of the magnetothermamal hysteresis observed in the magnetically ordered phase of the moderately mass-enhanced U intermetallic UPt$_2$Si$_2$. Aside from the antiferromagnetically ordered state below $T_N$=31 K, in a susceptibility study we establish the presence and field dependence of two irreversibility temperatures $T_{irr}$ and $T_{irr}$. Comparing the susceptibility data to the result of a neutron-scattering study, we find the lower temperature $T_{irr}$ to be accompanied by an anomaly in the temperature dependence of various magnetic Bragg peaks, viz., a reduction of the intensity. Tentatively, we attribute this behavior to a modification of the symmetry of the magnetically ordered phase. Altogether, our findings imply that the magnetic phase diagram of UPt$_2$Si$_2$ is much richer than previously thought, and similar to related uranium (122) intermetallic compounds. © 2006 American Institute of Physics. [DOI: 10.1063/1.2165917]

Crystallographic disorder can dramatically affect the properties of heavy fermion compounds.$^{1,2}$ Correspondingly, non-Fermi-liquid states, spin-glass behavior, or anomalous transport properties have been associated with the presence of disorder in materials such as UCu$_3$Pd, URh$_2$Ge$_2$, or U$_2$TSi$_3$.$^{3-5}$ Recently, based on a susceptibility study, we have presented evidence that also the moderately mass-enhanced (electronic specific-heat coefficient $\gamma=32$ mJ/mol K$^2$) uranium ternary UPt$_2$Si$_2$ is crystallographically disordered.$^6$

With respect of studying the effect of disorder on the physical properties of heavy fermion compounds, UPt$_2$Si$_2$ is a much more suitable material than the other systems studied so far. For UPt$_2$Si$_2$, large single crystals can be grown. Moreover, these crystals can be studied in close detail, in particular, by means of neutron scattering, since the scattering cross sections of U, Pt, and Si are quite different (U: 8.906 b, Pt: 11.71 b, and Si: 2.167 b), yielding a very bright elemental contrast and high sensitivity regarding disorder effects. Therefore, we have initiated an extensive investigation of UPt$_2$Si$_2$ by means of both thermodynamic/transport techniques and neutron scattering. In this contribution, in a first step, we correlate our observation of an anomalous magnetothermamal hysteresis in the susceptibility with the temperature dependence of the magnetic Bragg scattering component.

UPt$_2$Si$_2$ has been reported to crystallize in the tetragonal CaBe$_2$Ge$_2$ structure.$^{7,8}$ Further, the system undergoes an antiferromagnetic (AFM) transition at low temperatures, with $T_N$ determined to 38 (Ref. 7) and 35 K$^8$ respectively. From the appearance of magnetic Bragg intensity for the $[100]$, $[102]$, and $[111]$ peaks it was concluded that the magnetic structure consists of magnetic moments ($\mu_{\text{mag}}\sim 1.7\mu_B$) pointing along the $c$ axis, being ferromagnetically coupled within the $a$-$b$ plane and antiferromagnetically stacked along the $c$ axis. Unlike related uranium compounds such as UNi$_2$Si$_2$, UNi$_2$Ge$_2$, or UPd$_2$Si$_2$, where multiple magnetic phases have been observed, in UPt$_2$Si$_2$ the simple AFM structure was reported to persist down to lowest temperatures.

A more recent investigation indicated the occurrence of a first-order structural transition at $T_N$=305 K.$^{12}$ While the details of the structural transition have not been resolved, with a melting point above 1000 K a first-order phase transition at a comparatively low temperature likely leads to frozen-in disorder.

In fact, in a study on a newly prepared single-crystal UPt$_2$Si$_2$ (Ref. 6) we have observed an anomalous magnetothermamal hysteresis. In addition, by now we have carried out a full characterization of our as-cast crystal by means of bulk thermodynamic, transport, and microscopic techniques, about which a full account will be given elsewhere.$^{13}$ From a detailed structural single-crystal neutron-scattering study we find our sample to crystallize in the tetragonal CaBe$_2$Ge$_2$ lattice, in agreement with Refs. 7 and 8. Moreover, these experiments yield evidence for ligand disorder in the form of anomalously large thermal displacement parameters in the tetragonal plane for the Pt(2) and Si(2) sites in the CaBe$_2$Ge$_2$ structure. Essentially, it implies that in UPt$_2$Si$_2$ there is disorder in one-half of the unit cell in the form of a strained Pt/Si layer.

From our study of the bulk properties, in agreement with previous reports, the as-cast crystal undergoes an antiferromagnetic transition, although at a substantially lower tem-

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Neutron Scattering Center (BENSC) of the HMI, using the E1 spectrometer of the BENSC, with a neutron wavelength of $\lambda = 2.42757$ Å. We have measured the temperature evolution of the Bragg intensity for ten different peaks accessible in the geometry of the three-axis spectrometer. We find additional Bragg intensity in the magnetically ordered phase on the $[100]$, $[102]$, $[103]$, and $[201]$ peaks, while the other six peaks only show a constant nuclear scattering component. The appearance of magnetic Bragg intensity on these positions is fully consistent with the magnetic structure proposed in Ref. 8. The overall temperature dependence of the $[100]$ peak corresponds to that published in Ref. 8, but now with a transition temperature $T_N = 32.1$ K (Fig. 3, for a full account see Ref. 13).

Surprisingly, all four Bragg peaks reveal an intensity anomaly as temperature is lowered below $T_{irr2}$. This is illustrated in Fig. 3, where we plot the $T$ dependence of the Bragg intensity for these peaks at low temperatures. This anomaly, viz., a reduction of the scattering intensity below $T_{irr2}$, is most pronounced for the $[103]$ peak, where most of the magnetic intensity vanishes. It is somewhat weaker for the $[102]$ and $[201]$ peaks, and also for the $[100]$ reflection peak.

FIG. 1. The low-temperature susceptibility $\chi$ of UPt$_2$Si$_2$, measured along the $c$ axis in field-cooled (FC) and zero-field-cooled (ZFC) mode for various fields (upper panel), and the difference between FC and ZFC susceptibilities $\Delta \chi$, as determined from the field-dependent susceptibility data (lower panel). The lines indicate the extrapolation scheme to derive $T_{irr2}$ (for details see text).

FIG. 2. The magnetic phase diagram ($T_{irr}$, $T_N$, $T_{irr2}$) for fields $B$ directed along the $a$ and $c$ axes of UPt$_2$Si$_2$, as derived from the magnetic susceptibility in Fig. 1. The lines are guides to the eyes.

FIG. 3. The temperature dependence of the magnetic Bragg intensity for the $[100]$, $[102]$, $[103]$, and $[201]$ peaks of UPt$_2$Si$_2$ in the magnetically ordered phase. The lines are guides to the eyes.
we see some small intensity decrease. Thus, it appears that the anomaly is more pronounced for larger Bragg scattering angles.

At this point, we do not have a full explanation for the intensity anomaly of the magnetic Bragg peaks. A possible explanation for the intensity decrease, that is a reduction of the magnetic moment below \(T_{\text{irr2}}\), appears unlikely, as it should lead to an intensity reduction for all four peaks by the same factor. Further, since the [1 0 0] is the least affected by the anomaly, it implies that spin reorientation away from the \(c\) axis is minute. Conversely, the apparent angular dependence seems to indicate that at \(T_{\text{irr2}}\) changes of the form factor occur. However, again, such behavior would be very unusual, and it is not clear why this should happen in UPt\(_2\)Si\(_2\).

Instead, we speculate that the anomaly corresponds to a modification of the symmetry of the magnetically ordered state. Since the intensities are only changed, it indicates that the magnetic structures above and below \(T_{\text{irr2}}\) are closely related, and that there is not a complete symmetry change occurring at \(T_{\text{irr2}}\). In this situation, we would expect to see additional magnetic intensity coming up below \(T_{\text{irr2}}\) for some new Bragg peak positions. This additional intensity, however, must be quite small, as in the previous neutron powder-diffraction experiments in the difference spectra for no other Bragg peaks than the ones studied here magnetic intensity has been recorded. In analogy to related compounds,\(^9,10\) we speculate that a long-range modulation of the basic magnetic structure of UPt\(_2\)Si\(_2\) possibly might explain the observed behavior. Certainly, the magnetic phase diagram of UPt\(_2\)Si\(_2\) appears to be much richer than previously thought.

With respect to disorder, the two irreversibility temperatures reflect different aspects. The upper temperature, \(T_{\text{irr}}\), is the result of the strong dependence of the relevant energy scales on the local magnetic coupling strength \(J\). In a Doniach-like picture magnetic ordering results out of a competition between Kondo effect [energy scale \(T_K \propto \exp(-1/JN(E_F))]\) and the RKKY exchange [energy scale \(T_{\text{RKKY}} \propto J^2N(E_F)]\).\(^16\) For UPt\(_2\)Si\(_2\) we are in the localized limit of comparatively small \(J\) values, implying that the RKKY exchange wins and that the system undergoes a magnetic transition at \(T_N\). However, with the local variation of \(T_K\) and \(T_{\text{RKKY}}\) as result of the variation of \(J\), locally there are magnetically stronger coupled regions, which give rise to the inhomogeneous cluster state between \(T_N\) and \(T_{\text{irr}}\). Thus, \(T_{\text{irr}}\) directly reflects the disorder-induced variation of \(J\).

In contrast, \(T_{\text{irr2}}\) shows up in the FC/ZFC susceptibility as here the crystallographic disorder provides the centers for the antiferromagnetic domains to be pinned. If we assume that a modulation of the magnetically ordered structure occurs below \(T_{\text{irr2}}\), evidently the AFM domain structure will be different above and below \(T_N\). This difference is tested in the FC/ZFC susceptibility experiment, and is visible only because of the presence of pinning centers.

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\(^{12}\)H. Amitsuka (private communication).
\(^{15}\)The peaks searched for magnetic Bragg intensity are [1 0 0], [0 0 1], [1 0 1], [2 0 0], [0 0 2], [1 0 2], [2 0 1], [0 0 3], [1 0 3], and [0 0 4].