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# Structural properties, magnetic order and electronic transport in single crystalline $UPt_2Si_2$

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ABSTRACT

We present a detailed comparison of the physical properties of as-cast and annealed single crystalline $UPt_2Si_2$, a compound whose properties we have shown to be governed by strain disorder on the Pt/Si ligand sites. Contrary to common knowledge, and to our surprise, from our data we do not observe a significant improvement of the physical properties of $UPt_2Si_2$ upon annealing at 900 °C for one week. We attribute this to the specific way the strain disorder is produced in $UPt_2Si_2$ by presenting evidence that it results from a first order phase transition at ambient temperatures. We discuss the implications of such phase transitions occurring at comparatively low temperatures for the ground state properties of heavy fermion systems and related correlated electron materials.



## 1. Introduction

The effect of structural disorder on the physical properties of strongly correlated electron systems is a topic of current interest in this field of condensed matter physics [1–6]. In particular, for heavy fermion systems crystallographic disorder is known to affect the magnetic ground state properties [7]. Moreover, disorder has been proposed to significantly affect the physics close to magnetic instabilities, with for instance non-Fermi liquid behavior occurring in the vicinity of a quantum critical point [8–14].

In this situation, in order to better understand and control disorder effects in heavy fermion materials, two different approaches have been chosen in corresponding studies in recent years. First, various heavy fermion materials have been identified where crystallographic disorder does affect the physical properties. These systems have been studied intensively, in order to characterize on a qualitative and quantitative level how and to what degree the disorder controls the physics of heavy fermions [7,8,15–35].

Secondly, by subsequently improving the process of material production, handling and characterization an attempt is made to identify the intrinsic properties of the "perfect" compound [36–43]. For this, preferably single crystalline materials should be studied. Further, heat treating the material is a common procedure to reduce the level of structural disorder in crystalline materials [13,22,44]. This approach assumes that disorder is the result of a comparatively fast cooling procedure during material production causing local strain or stoichiometry fluctuations, both which can be removed through annealing.

However, in some cases crystallographic disorder may be the result of a structural instability, and which will not be removed through annealing. Here, we will discuss one such case, namely the ternary uranium compound $UPt_2Si_2$, a heavy fermion like intermetallic with a moderately enhanced Sommerfeld coefficient to the specific heat either reported as $\gamma = 32\,\mathrm{mJ/mole\,K^2}$ or $122\,\mathrm{mJ/mole\,K^2}$ [45]. For this material, we compare the physical behavior for as-cast and annealed single crystalline material. Our data indicate that the atomic scale strain disorder present at low temperatures is inherent to this compound, and most likely results from a first order phase transition occurring slightly above room temperature. A review of literature indicates that such first order phase transition induced disorder is a quite common occurrence.

Ternary silicides of composition (Ce,U)$T_2Si_2$, $T$=transition metal, crystallizing either in the tetragonal $ThCr_2Si_2$ (space group $I\,4/mmm$) or the $CaBe_2Ge_2$ ($P\,4/nmm$) structure, exhibit a rich variety of physical phenomena, such as heavy fermion behavior, local moment antiferromagnetism, or hidden order phases in combination with heavy fermion superconductivity [45–48].

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Inherent to this class of compounds is a competition between the $ThCr_2Si_2$ and the $CaBe_2Ge_2$ lattice. For instance, those systems $UT_2Si_2$ with a light *d* element ($T$=Cr, Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd) [49–55] crystallize in the $ThCr_2Si_2$ structure, while those compounds containing heavy elements ($T$=Ir, Pt) have been reported to form in the $CaBe_2Ge_2$ lattice [56,57]. For "intermediate" cases such as the Au and Os containing compounds their structural properties have not been established definitely [58–60].

Some of the compounds (Ce,U)$T_2Si_2$ even exhibit transitions from the one to the other crystal structure. As an example, as-cast fast cooled $CeIr_2Si_2$ crystallizes in the $CaBe_2Ge_2$ structure with almost tetra-valency of the Ce ion, while an annealing at 600 °C transforms the compound into the $ThCr_2Si_2$-type structure with a magnetic behavior of a mixed valent system [61]. Quite similarly, the very different magnetic properties of various samples $UCo_2Ge_2$ are caused by the two different crystal structures, being antiferromagnetic for annealed material crystallizing in the $ThCr_2Si_2$ lattice and non-magnetic for as-cast samples with the $CaBe_2Ge_2$ symmetry [62].

$UPt_2Si_2$ belongs to those 122-compounds which crystallize in the $CaBe_2Ge_2$ lattice [45,57,63]. However, frozen-in structural disorder is present in $UPt_2Si_2$, and which controls many of its physical properties [63]. The cause for the occurrence of the disorder has not yet been identified conclusively. In view of the published results on other 122 cerium or uranium intermetallics, in order to fully understand $UPt_2Si_2$ one route of investigations is to study the influence of an annealing process on the crystallographic structure and the physical properties. From such a study a better understanding of what is causing the atomic scale disorder in $UPt_2Si_2$ and related intermetallics can be obtained.

In order to achieve this goal, this paper is organized as follows. First, we present a detailed comparison of the properties of as-cast (ac) and annealed (ann) single crystalline $UPt_2Si_2$. We have performed a detailed structural investigation, in order to determine if the level of crystallographic disorder can be reduced by annealing. From our neutron scattering study we detect frozen-in strain disorder on two of the four different ligand (Pt/Si) sites for annealed $UPt_2Si_2$, very similar to the situation for the as-cast material. Regarding the ground state properties of $UPt_2Si_2$, they are not significantly influenced by annealing, in stark contrast to the expectation of an improved sample quality. In view of these results we present additional experiments of various bulk properties on single crystalline $UPt_2Si_2$, which have been carried out to above room temperature and provide evidence for a first order phase transition occurring at about 305 K. We believe that this phase transition can provide the clue to understand the presence of atomic scale disorder in $UPt_2Si_2$, as it might produce the strained regions observed experimentally.

## 2. Results and discussion

For our annealing study we have used the tri-arc Czochralski grown single crystal $UPt_2Si_2$ previously studied in Ref. [63] and labeled crystal ♯1 therein. After the experiments described in this reference have been carried out, several pieces of the crystal were wrapped in Ta foil and annealed in an evacuated quartz ampoule at 900 °C for one week. Subsequently, these pieces were investigated regarding their electronic transport properties and susceptibility. Resistivity measurements were carried out by means of a standard ac four-point technique at temperatures ranging from 2 to 300 K. The susceptibility was measured employing a commercial SQUID magnetometer, at temperatures ranging from 5 to 300 K. Further, single crystal neutron diffraction studies have been performed at the Berlin Neutron Scattering Center (BENSC) of the Helmholtz–Zentrum Berlin using the diffractometers E1 (neutron wave length $\lambda = 2.42757$ Å) and E5 (neutron wave length $\lambda = 2.4307$ Å), to establish the structural properties, the level of structural disorder and the characteristics of the antiferromagnetic phase. Based on the results from these studies, we have initiated additional resistivity and susceptibility experiments on the tri-arc Czochralski grown single crystal $UPt_2Si_2$ labeled crystal ♯2 in Ref. [63], but now up to temperatures (far) above room temperature.

For the study of the structural properties of $UPt_2Si_2$ we have carried out extensive neutron scattering experiments on the annealed crystal analogous to those performed on the as-cast one. Thus, for the structure refinement we have used the E5 diffractometer at BENSC at a low temperature of 50 K. As reported previously [45,51,63] we find that the system crystallizes in the $CaBe_2Ge_2$ lattice (space group $P\,4/nmm$). For the refinement of the data from the annealed crystal we have left all parameters, including the site occupation, free. With respect to the site occupation, from our analysis we find as best structural solution a fully ordered lattice, i.e., within experimental error ($\pm 3\%$) there is no evidence for random site exchange in the annealed material, in close agreement with results obtained for the as-cast crystal.

In Table 1 we summarize the result of an optimum refinement of our data. In the table we compare the results with those previously obtained for the as-cast crystal [63]. First of all, from this comparison it can be seen that regarding lattice parameters, the free *z* positions and quality of the fit there are no significant differences between the two sets of data.

In contrast, and quite surprisingly, the values for the displacement parameters $U_{11}$ and $U_{33}$ are larger (although not significantly) for the annealed sample than for the as-cast one. Moreover, it can be seen that for both the annealed and the as-cast material the values for the displacement parameter $U_{11}$ for the Pt(2) and Si(2) sites are extraordinarily large, if compared to the other sites in this lattice. As we have demonstrated previously

**Table 1**
The result of refinements of the single crystal neutron diffraction data on as-cast (*ac*) and annealed (*ac*) $UPt_2Si_2$, with the $CaBe_2Ge_2$ lattice (space group $P\,4/nmm$).

| | *x* | *y* | *z* | $U_{11}$ (Å$^2$/8π$^2$) | $U_{33}$ (Å$^2$/8π$^2$) |
|---|---|---|---|---|---|
| U | | | | | |
| *ac* | 1/4 | 1/4 | 0.7484(2) | 0.45(4) | 0.08(4) |
| *ann* | 1/4 | 1/4 | 0.7480(2) | 0.66(5) | 0.26(7) |
| Pt(1) | | | | | |
| *ac* | 1/4 | 1/4 | 0.3785(2) | 0.10(3) | 0.28(4) |
| *ann* | 1/4 | 1/4 | 0.3787(2) | 0.25(5) | 0.39(6) |
| Pt(2) | | | | | |
| *ac* | 3/4 | 3/4 | 0 | 1.86(5) | 0.14(4) |
| *ann* | 3/4 | 3/4 | 0 | 1.98(6) | 0.30(5) |
| Si(1) | | | | | |
| *ac* | 3/4 | 1/4 | 1/2 | 0.10(6) | 0.18(7) |
| *ann* | 3/4 | 1/4 | 1/2 | 0.16(7) | 0.38(9) |
| Si(2) | | | | | |
| *ac* | 1/4 | 1/4 | 0.1330(3) | 0.94(8) | 0.29(8) |
| *ann* | 1/4 | 1/4 | 0.1340(4) | 1.27(10) | 0.42(12) |

Lattice parameters/refinement value
As-cast: $a = 4.186$ Å; $c = 9.630$ Å/$R_{Bragg} = 6.6\%$
Annealed: $a = 4.186$ Å; $c = 9.630$ Å/$R_{Bragg} = 6.6\%$

Experiments have been carried out at 50 K.