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On some magnetic phase transitions in CePtSn

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Abstract

We present measurements of the magnetization and specific heat of a CePtSn single crystal in the temperature range 1.7–15 K and in magnetic fields up to 14 T applied along the orthorhombic *b* axis. Possible microscopic mechanisms responsible for the observed magnetic phase transitions are discussed in the context of previously reported magnetoresistance and neutron-scattering data. Special emphasis is put on an irreversible field-induced transition around 4 T observed for temperatures $T \leq 2$ K. At this transition, found when sweeping the field up from the zero-field-cooled (ZFC) state, a tiny change in the magnetization is accompanied by a large negative GMR effect. Surprisingly, no transition is observed when sweeping the field back to zero, i.e. the 'field-annealed' phase (after the application of a sufficiently high field) persists even in zero field and a simple AF structure with a propagation vector $\boldsymbol{q}=(0,1/2,0)$ is proposed to characterize this phase. Clear evidence for this transition is observed also in temperature dependencies of the magnetization and specific heat in various magnetic fields. Based on these results, the completed magnetic-phase diagram of CePtSn in fields parallel to the *b* axis is presented.

Keywords: Intermetallics; Magnetization; Specific heat; Anisotropy

1. Introduction

CePtSn crystallizes in the orthorhombic ε-TiNiSi-type structure. According to the literature [1,2] this dense Kondo compound orders antiferromagnetically (AF) below $T_N=7.5$ K and undergoes a second magnetic transition at $T_M=5$ K. The latter transition was reported to be of the first-order [2]. Neutron-diffraction studies by Kadowaki et al. [3] on a different single crystal were initially interpreted in terms of two incommensurate AF structures of Ce magnetic moments characterized by the propagation vectors $\boldsymbol{q}=(0,\delta,0)$ with $\delta=0.466$ and 0.418 for temperatures $T<T_M$ and $T_M<T<T_N$, respectively. On the other hand Kolenda et al. reported a considerably different value of $\delta=0.428$ obtained from neutron diffraction on a powder CePtSn sample at 1.7 K [4]. The onset of magnetic ordering in CePtSn was also confirmed by NMR experiments on the ^{119}Sn and ^{195}Pt nuclei that revealed the NMR signals to disappear below 7 K [5]

Later on, Kalvius et al. [6] performed μSR experiments on CePtSn and found that their results were inconsistent with the incommensurate structures proposed from neutron diffraction results. Neither of the magnetic phases show a broad frequency distribution as expected for truly incommensurate structures, but one finds a single-frequency μSR spectrum for temperatures $T_M<T<T_N$ and three discrete frequencies below T_M.

To resolve the contradiction between the neutron and μSR data, Kadowaki has proposed a spin-slip model based on the irreducible representation analysis [7,8]. He pointed out that the $\boldsymbol{q}$ appears to be incommensurate for both phases only due to periodical occurrence (with the period of ~20 lattice spacings) of spin-slip planes within the essentially commensurate phase with $\boldsymbol{q}=(0,1/2,0)$.

Magnetization measurements performed on CePtSn single crystals reveal strong magnetocrystalline anisotropy with the easy-magnetization direction along the *a* axis, which can be attributed to the crystal–field interaction [2]. A set of reliable crystal–field coefficients was derived by Divis et al. [9] which accounts for the available susceptibility, high-field magnetization [2] and inelastic-neutron-scattering data [10,11].

Previous high-field magnetization measurements [2] at 1.5 K for fields applied along the *b* axis revealed a small step-like metamagnetic transition near 11 T with some hysteresis. This transition disappears with increasing tem-

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perature above 3 K. Recent magnetoresistance measurements at 1.8 K revealed that this transition is accompanied by a considerable positive magnetoresistance step [12]. The related magnetization data at 2.5 K presented in the same paper show the transition at a practically identical field. In addition to this anomaly, striking resistivity behavior is observed at lower fields. Initially, the *b* axis magnetoresistance exhibits parabolic increase with field up to 3.5 T where it reaches ~1.03 of the zero-field value.

A continuous parabolic increase of the resistance with applied field is reminiscent of normal magnetoresistance in a metal. With further increasing the field, the resistance drops suddenly around 4 T by ~35% although no sign of transition is seen on the 2.5-K magnetization curve. Between 4 and 11 T, the resistance increases continuously again. When sweeping the field down from 18 T, the '11 T' transition is reversible, but, surprisingly, the '4 T' transition does not show up and the final zero-field resistivity value is reduced by 36% with respect to its initial value.

Here, we report on detailed magnetization and specific-heat measurements performed on a CePtSn single crystal at temperatures from 1.7 to 15 K and in magnetic fields up to 14 T applied along the *b* axis. The main motivation of the present study is to see whether the irreversible magneto-resistance effect bellow 4 T is connected with magnetism in CePtSn.

2. Experimental

A single crystal of CePtSn was grown under Ar atmosphere by a modified Czochralski method using a tri-arc furnace using stoichiometric amounts of starting materials (Ce, 4 N; Pt, 3 N; Sn, 5 N). The quality of the crystal was checked by X-rays and thermal neutrons (at ILL, Grenoble). For specific-heat measurements, a 20-mg platelet with faces perpendicular to the *b* axis was cut by spark erosion from the main crystal. Similarly, an ~8-mm^3 rectangular-shaped sample with faces parallel to the three principal directions was cut for magnetic measurements. Lower-field magnetic measurements were performed on a Quantum Design SQUID magnetometer in fields up to 5 T. Our results for the temperature dependence of magnetic susceptibility (4.5–300 K) and the low-temperature magnetization curves (on zero-field-cooled (ZFC) samples) for fields along the three principal directions are in very good agreement with previously published data [2].

Specific heat and magnetization were measured at temperatures from 1.7 to 15 K and magnetic fields up to 14 T applied along the *b* axis using a physical property measuring system (PPMS) from Quantum Design that is equipped with a 14-T superconducting coil. A relaxation method was used for specific-heat measurements. Magnetization was measured using an extraction-magnetometer insert sweeping the field from 0 to 14 T and back to zero field. Each measurement of specific heat and magnetization began from the ZFC state.

3. Results and discussion

Fig. 1 shows the magnetization curves in fields up to 14 T measured on the CePtSn crystal at 2, 3, 4, 5 and 6 K oriented with *b* axis in the field direction. The *M(B)* curve at 2 K exhibits a hysteretic step-like transition at 11 T in excellent agreement with previous data [2]. Already at slightly elevated temperatures, a very different magnetic response is found. The hysteresis vanishes for temperatures above 3 K, and (instead of a step at 11 T) a broad S-shaped magnetization curve between 5 and 11 T (above which the *M* vs. *B* is linear) is seen. This transition is pushed down to 8.5 T at 4 K, and no S-shape can be distinguished for temperatures above 5 K, where the

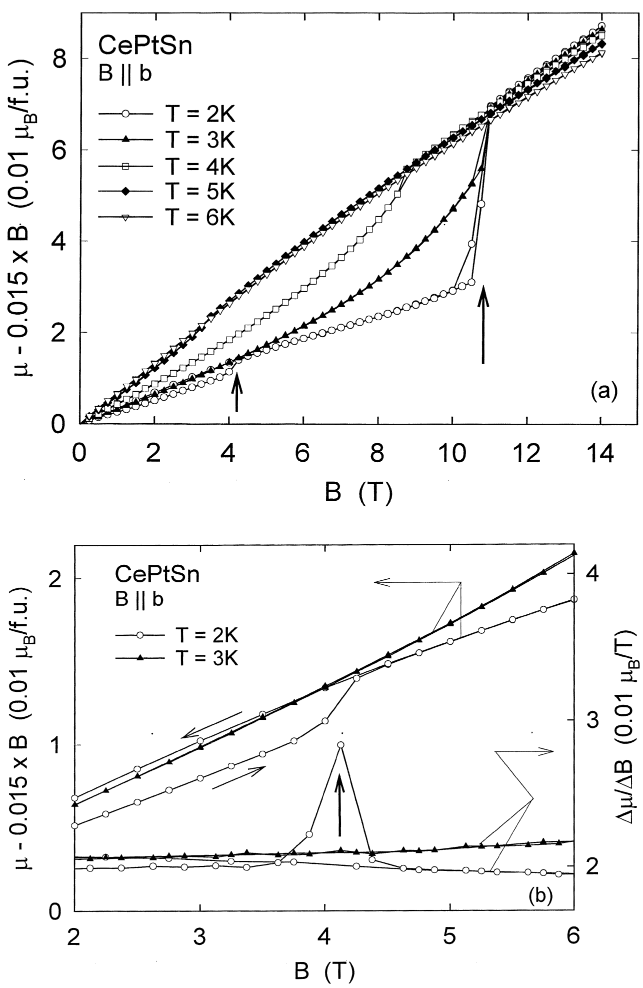

Fig. 1. (a) Field dependence of the magnetization of CePtSn measured at T=2, 3, 4, 5 and 6 K in fields up to 14 T applied along the *b* axis. (b) Detailed behavior of the magnetization and differential susceptibility around 4 T at T=2 and 3 K. The slope of magnetization curves is artificially reduced by 0.015 to visualize the transitions better. The lines are guides for the eye.

magnetization varies almost linearly with field with some small saturation tendency at higher fields.

Another unusual feature on the 2-K magnetization curve can be seen around 4 T. For the sake of clarity, the region around 4 T is displayed in detail in Fig. 1b. One observes some tiny step in the magnetization when sweeping the field up across 4 T from the ZFC state. A large negative giant magnetoresistance (GMR) effect was reported previously at about the same field [12]. Similar to the magnetoresistance observation, we find that this transition is absent when sweeping the field back to zero indicating that the 'field-annealed' phase persists to 0 T.

The huge reduction of resistance at the '4 T' transition may reflect a considerable change of magnetic periodicity while preserving antiferromagnetic ordering. Then, considering the spin-slip model presented for CePtSn by Kadowaki [7,8], we propose the following scenario for the '4 T' transition. The ZFC state for $T \leq 2$ K is characterized by a spin-slip phase that can be viewed as a defected $\boldsymbol{q}=(0,1/2,0)$ structure with defects (spin-slips) appearing periodically along the b axis over ~20b. For magnetic fields higher than 4 T, the defects (spin-slips) are driven out of the sample yielding a simple $\boldsymbol{q}=(0,1/2,0)$ propagation of Ce moments. When the field is removed, the spin-slip phase is not recovered and we are left with a simple $\boldsymbol{q}=(0,1/2,0)$ antiferromagnet.

On the other hand, the '11 T' transition at 2 K may reflect a 'real' change of the magnetic structure and a subsequent change of the magnetic periodicity. In view of the sudden magnetization step at this transition, the low-temperature ($T<3$ K) high-field ($B>11$ T) phase may be then speculated to be an uncompensated antiferromagnetic phase (UAF). Neutron-diffraction experiments in magnetic fields should be carried out to validate the above scenario for the b axis transitions observed in CePtSn.

The temperature dependencies of the magnetization and specific heat of CePtSn in magnetic fields up to 14 T applied parallel to b axis are displayed in Figs. 2 and 3, respectively. The existence of the '4 T' transition is confirmed by these measurements. In particular, anomalies are observed in the vicinity of 3 K in *M(T)* and *C(T)* data in fields ≥4 T for heating from the 2-K ZFC state, whereas there is no anomaly seen in lower fields. The '11 T' transition is reflected in the dramatic change between the low-temperature *M(T)* and *C(T)* behavior for fields lower and higher than the transition field.

The anomalies in *M(T)*, *M(B)* and *C(T)* allowed us to construct the magnetic phase diagram of CePtSn for fields applied along the b axis (Fig. 4). We have confirmed the previously proposed phase boundaries [12], but in addition we identified new boundaries and new phases that are closely connected with the '4 T' and 11 T metamagnetic transitions at temperatures below 3 K and by transitions indicated around 3 K in *M(T)* and *C(T)* data at various fields ≥4 T. Further details are given in the caption of Fig. 4. It should be noted that some evidence of the irreversible

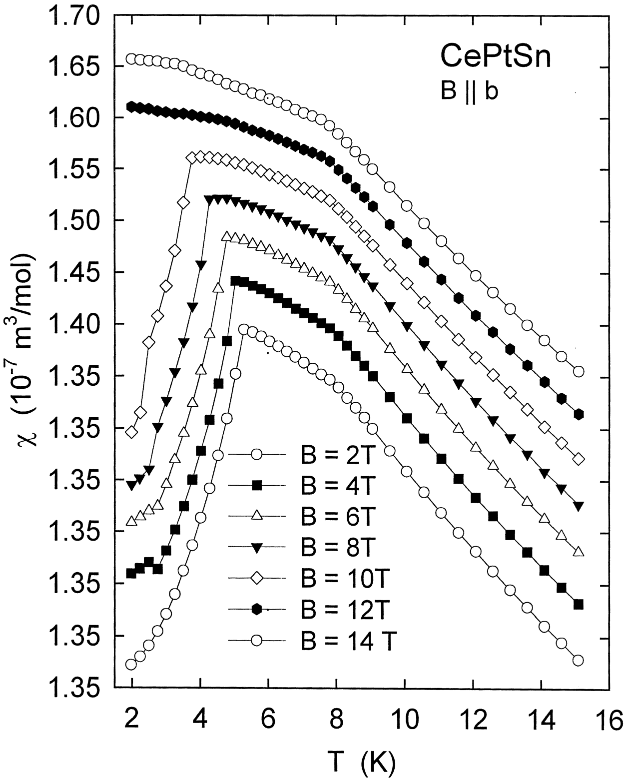

Fig. 2. Temperature dependence (for $T \leq 15$ K) of the susceptibility of CePtSn ($\chi = M/B$) in magnetic fields up to 14 T applied along the b axis. The lines are guides for the eye.

'4 T' transition has been detected already in pulsed magnetization measurements at 1.5 K and in specific-heat data in 5 T published in Ref. [2]. The state of the art at that time prevented any conclusion being made. Intensive neutron diffraction study in various magnetic fields is necessary to determine various microscopic aspects of magnetic ordering in CePtSn.

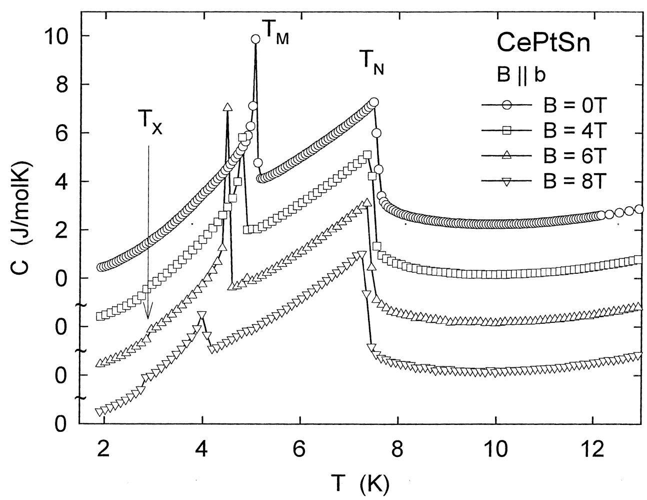

Fig. 3. Temperature dependence (for $T \leq 13$ K) of the specific heat of CePtSn in magnetic fields up to 8 T applied along the b axis. The lines are guides for the eye.

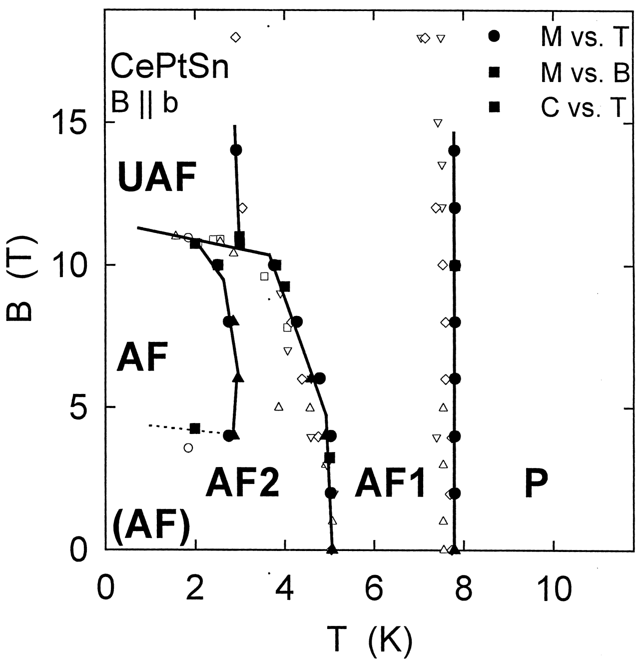

Fig. 4. Schematic drawing of the magnetic phase diagram of CePtSn in magnetic fields up to 14 T applied along the *b* axis. The open symbols are taken from Ref. [1] (△) and Ref. [12]. The solid symbols originate from our measurements of *M* versus *B* (■), *M* versus *T* (●) and *C* versus *T* (▲) dependencies. The lines are guides for the eye. The proposed phases are AF1 and AF2 analogous to those presented in Ref. [12] but with a limited stability *B*–*T* space. AF2 is observed only in the ZFC state whereas the AF phase is characterized by the low resistance most probably with $\boldsymbol{q}$=(0,1/2,0). The AF phase is found only at temperatures below 3 K and after exerting the crystal to fields above 4 T. The UAF phase characterized by high magnetization and high resistivity is confined in the section of the *B*–*T* diagram for $B>11$ T and $T<3$ K. In our scenario this phase is most probably an uncompensated antiferromagnetic phase.

In conclusion, we have measured magnetization and specific heat on a single crystal of CePtSn with respect to temperature and magnetic field applied along the *b* axis. In zero fields, the existence of two magnetic transitions was confirmed with the characteristic temperatures T_N=7.5 K and T_M=5 K, respectively. The low-temperature magnetization curves exhibit two metamagnetic transitions around 4 and 11 T, respectively. The lower transition is observed to be irreversible which is consistent with previous magnetoresistance data [12]. A microscopic scenario was discussed allowing understanding of the peculiar magnetic phase transitions. Neutron-diffraction experiment on an identical single crystal in magnetic fields is scheduled to test the proposed picture.

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