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Crystal growth and magnetic property of a new compound CeAu₄Si₂

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Abstract

We have succeeded in growing a new ternary Ce compound CeAu₄Si₂. This compound crystallizes in the tetragonal crystal structure $P\bar{4}m2(\sharp 115)$ with lattice parameters a = 4.3304 Å and c = 27.409 Å. The local environment around Ce atoms is similar to that of the well-known ThCr₂Si₂-type tetragonal structure in CeAu₂Si₂. Single crystals of CeAu₄Si₂ were grown by the flux method using an Au_{4.37}Si eutectic alloy as flux. Electrical resistivity, specific heat and magnetization measurements revealed a ferromagnetic ordering at $T_{\rm C} = 5.3$ K with a saturation moment of $1.7 \mu_{\rm B}/{\rm Ce}$. Anisotropic features in the magnetic susceptibility and magnetization were well explained by a crystalline electric field (CEF) model. Estimated excitation energies from the ground state doublet to two excited doublets are 284 and 340 K, respectively.

Keywords: Rare-earth alloys and compounds; Intermetallics; Crystal growth; Magnetization

1. Introduction

The 4f electrons of rare-earth compounds exhibit a variety of characteristic features including spin and valence fluctuations, spin and charge orderings, heavy fermions and anisotropic superconductivity [1]. Two competing interactions, namely the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect, play a pivotal role in the observed magnetic properties. The RKKY interaction produces the long-range magnetic order, where the 4f electrons with the magnetic moments are treated as the localized electrons, and the indirect f-f interaction is mediated by the spin polarization of the conduction electrons. On the other hand, the Kondo effect quenches the magnetic moments of the localized 4f electrons by the spin polarization of the conduction electrons, producing a singlet state with the binding energy $k_{\rm B}T_{\rm K}$, where $T_{\rm K}$ is called the Kondo temperature, which leads to the heavy-fermion state with an extremely large effective mass at temperatures lower than $T_{\rm K}$.

The competition between the RKKY interaction and the Kondo effect was discussed by Doniach [2] as a function of $|J_{cf}|D(\epsilon_{\rm F})$, where $|J_{cf}|$ is the magnitude of the magnetic ex-

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change interaction and $D(\epsilon_{\rm F})$ is the electronic density of state at the Fermi energy $\epsilon_{\rm F}$. Most of the cerium compounds order magnetically, because the RKKY interaction overcomes the Kondo effect at low temperatures. On the other hand, some cerium compounds such as CeCu₆ and CeRu₂Si₂ exhibit no long-range magnetic order.

In recent days, there has been a considerable interest in the studies of cerium compounds near the quantum critical point where the electronic state reorganizes itself into a new stable phase of matter [1]. Our recent high pressure investigations on the ternary germanides, namely CeNiGe₃ and Ce₂Ni₃Ge₅, have clearly elucidated the existence of new electronic states around the quantum critical point, where the Néel temperature T_N becomes zero at a critical pressure $P_c : T_N \rightarrow 0$ at $P \rightarrow P_c$ [3,4]. The antiferromagnetic compound CeNiGe₃ with the orthorhombic crystal structure is found to become superconductive below 0.48 K in a wide pressure range from 4 to 10 GPa, and also in Ce₂Ni₃Ge₅ the electronic state is also found to be tuned by pressure for temperatures less than 0.26 K into a new stable superconducting phase around a critical pressure $P_c \simeq 3.9$ GPa.

Similar to the germanides, the rare-earth ternary intermetallic silicides also show interesting physical properties. The prototype heavy-fermion superconductor $CeCu_2Si_2$ [5], the pressureinduced superconductor $CePd_2Si_2$ [6] and the nonmagnetic

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heavy-fermion compound CeRu₂Si₂ [7] are some of the prominent examples of the rare-earth ternary silicides. Furthermore, very recently the coexistence of magnetism and superconductivity was reported for a heavy-fermion compound CePt₃Si with the tetragonal crystal structure [8]. One of the interesting features in CePt₃Si is that the crystal structure does not have a center of inversion. This compound exhibits an antiferromagnetic ordering at $T_N = 2.2$ K and is condensed into a heavy-fermion superconducting state for temperatures less than 0.75 K. From these results, it is expected that a novel superconducting state is realized in CePt₃Si.

In continuation to our studies on the germanides, we have synthesized a new ternary silicide, namely CeAu₄Si₂, both in polycrystalline and single crystalline forms. In this paper we present the structural and anisotropic magnetic properties of this new compound.

2. Experimental

We have prepared both the polycrystal and single crystal samples of CeAu₄Si₂. At first a polycrystalline alloy button of CeAu₄Si₂ was prepared by arc-melting the high pure metals of Ce(3N), Au(4N) and Si(5N) in the stoichiometric ratio in a tetra-arc furnace. The arc-melted button was then wrapped in a Ta-foil and subsequently annealed at 800 °C for 1 week in an evacuated quartz ampoule. This stoichiometric alloy button was used as the source material for the single crystal growth and also used as a reference material for comparison of the X-ray diffraction pattern between the polycrystal and single crystal samples.

Single crystals of CeAu₄Si₂ were grown by the flux method. Although the constituent metals have high melting points, the single crystals of this compound can be grown by the self-flux method. It is well known that the Au-Si system forms a deep binary eutectic at the composition 81:19 (Au:Si) with a eutectic temperature being 363 °C. In order to use it as a flux for the growth of CeAu₄Si₂ single crystals, we have taken advantage of this eutectic temperature and made an alloy button of Au-Si with the above composition by arc-melting. Appropriate amounts of this binary alloy, together with the polycrystal sample of CeAu₄Si₂, were taken in an alumina crucible. The crucible was then sealed in a quartz ampoule with a partial pressure of argon gas and placed inside an electric furnace. The temperature of the furnace was raised up to 1050 °C and the furnace was maintained at this temperature for 48 h in order to achieve proper homogenization of the melt. The furnace temperature was then slowly cooled down to 650 °C over a period of 2 weeks and finally cooled down to room temperature at a faster rate. Owing to the low melting point of the binary alloy (Au-Si), which was used as flux for the single crystal growth, it was removed by means of centrifuging. Large shining single crystals with dimensions $8 \text{ mm} \times 5 \text{ mm} \times 2 \text{ mm}$ were obtained, as shown in Fig. 1. The plane of the as-grown crystal was found to be perpendicular to the [001] direction, namely the (001) plane. The crystals were soft and malleable. The single crystals were then oriented by means of the back reflection Laue method and cut along the principal directions for the anisotropic physical property measurements.

The composition analysis of the as-grown single crystal and the polycrystal samples were examined using the electron-probe microanalysis (EPMA). The electrical resistivity along the principal directions was measured in the temperature range from 1.5 to 300 K using the standard dc four-probe method. The dc magnetic susceptibility and magnetization were measured in a commercial SQUID magnetome-



Fig. 1. As-grown single crystals of CeAu₄Si₂.

ter. The specific heat measurement down to 0.6 K was performed in a home-made calorimeter, which employs the quasi-adiabatic heat pulse method.

3. Experimental results and analyses

3.1. Composition and structural analyses

We analyzed the composition of both the as-grown single crystal and the polycrystal samples by EPMA. The analysis clearly indicated that both samples are almost homogenous and single phase. In the above analysis, we used the polycrystal sample as a reference to compare with the single crystals. The results of the composition analysis showed that Ce, Au and Si are in the ratio 1:4:2.

Next, we analyzed the crystal structure of this new compound CeAu₄Si₂. The single crystals were investigated by the back reflection Laue method. The Laue pattern revealed clearly the tetragonal symmetry with the Laue class 4/mmm. The as-grown plane of the single crystal corresponded to the (001) plane of the tetragonal cell. Both the single crystal and polycrystal samples were crushed into very fine powder and analyzed by the powder X-ray diffraction using a Cu Ka radiation. Powder spectra taken on both samples were essentially the same, although we observed a contamination of Au in the diffraction data of the flux-grown sample, which might be introduced from the flux. We therefore show in Fig. 2 the result taken for the polycrystal sample. The diffraction data were successfully indexed to the tetragonal unit cell, as shown in Fig. 2. Diffraction peaks due to impurity phases were not observed for the polycrystal sample. From the reflection indices and the structural consideration shown below, we determined that the space group is $P\bar{4}m2(\sharp 1\,1\,5)$ with the lattice parameters a = 4.3304(4) Å and c = 27.409(4) Å, which are compared with those of the wellknown ThCr₂Si₂-type tetragonal structure in CeAu₂Si₂ with the lattice parameters a = 4.301 Å and c = 10.21 Å.

In Fig. 3 we show the crystal structure of $CeAu_4Si_2$. The present structural model consists of three $CeSi_2$ layers denoted as A, A' and A'', and a double-Au layer denoted as B, as shown in Fig. 3. The layer B is very similar to the fcc Au-metal structure, while the arrangement of the layer A and the adjacent Aulayer is similar to that observed in the ThCr₂Si₂-type tetragonal CeAu₂Si₂. It is interesting to note that the layer A is shifted by Download English Version:

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