Magnetic structure of the local-moment antiferromagnet CeCuSn

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We report on single-crystal neutron diffraction studies of the local-moment antiferromagnet CeCuSn. The ground-state magnetic structure is characterized by a magnetic wave vector **k** $=(0.115,0,0)$. The onset of antiferromagnetic order occurs around 12 K with an inflection in the temperature dependence of the magnetic intensities at about 8 K. This is in contrast to bulk measurements, which only show broad features at 8–10 K. The ordered moments are likely reduced from the free-ion moment for Ce. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1850402]

Ce*T*Sn (T=transition metal) stannides have received considerable attention over the past decade due to their unusual ground-state properties. In particular, the orthorhombic compounds CeNiSn:[1] a Kondo semi-metal, and CePtSn,[2] which exhibits exotic magnetic order with irreversible metamagnetic transitions, have been the subjects of intense study. Fewer studies of the hexagonal Ce*T*Sn (T=Ag, Au, Sn) compounds have been conducted, perhaps partly because of their characterization as local-moment magnets, but also perhaps because single-crystal samples have not been available.

CeCuSn was reported to order antiferromagnetically below $T_N \approx 8.6$ K.[3–5] In addition, a broad double-peak feature in the zero-field specific heat was taken to indicate a second magnetic transition at about 7.5 K.[3] Subsequent muon spin relaxation (μSR) experiments indicated an unusual evolution of the magnetic ordering in CeCuSn, starting with the onset of short-range correlations at 11 K leading to spin freezing around 8.6 K, and finally a coexistence of long-range ordered and spin-frozen phases below 7.5 K.[6]

Recently, we synthesized single crystals of CeCuSn, allowing us to investigate the magnetic structure in greater detail. We were particularly motivated to perform single-crystal neutron diffraction experiments since very little information has been available regarding the ground-state magnetic structure of CeCuSn.

Single-crystalline samples of CeCuSn were grown from a ternary liquid.[7,8] The crystals have a hexagonal tabular habit, with clearly defined facets that coincide with the main crystallographic directions. The crystal structure of CeCuSn has been reported to be of the hexagonal $CaIn_2$ type (space group $P6_3/mmc$).[9] In this structure, the Cu and Sn atoms are distributed randomly on a single crystallographic site. However, it has also been suggested that CeCuSn may form in an ordered variant, the GaGeLi-type structure (space group $P6_3mc$), which is shown schematically in Fig. 1.[3,5] Single-crystal x-ray diffraction showed that our CeCuSn samples form in the hexagonal GaGeLi-type crystal structure with lattice parameters a=4.59 Å and c=7.90 Å.[10]

Bulk measurements on our samples are generally consistent with previously reported data for polycrystalline samples.[3–5] Magnetic order is accompanied by a slow upturn in the resistivity below about 10 K, and broad peaks in the temperature dependence of the magnetization and specific heat at about 8 K. However, it should be noted that all the features in our measurements were very broad, making it difficult to assign a Néel temperature. Furthermore, the magnetization is highly anisotropic, with the response to a magnetic field of the [001] direction magnetization much smaller than that perpendicular to it.

Neutron diffraction experiments were performed at the High Flux Isotope Reactor, Oak Ridge National Laboratory,

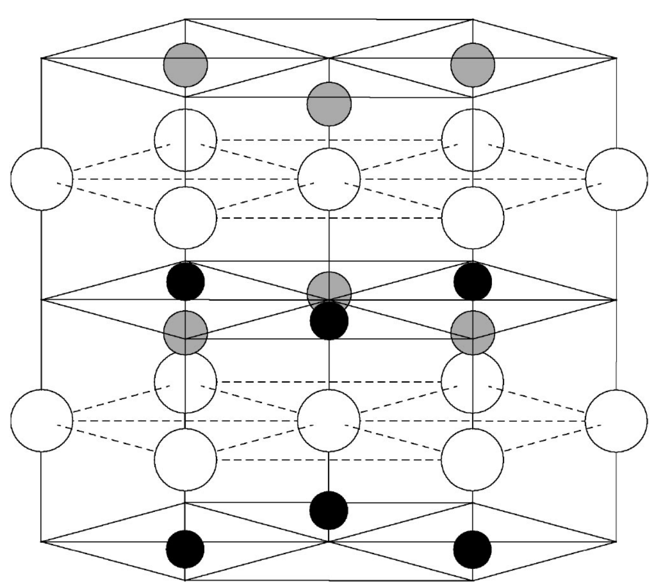

FIG. 1. Schematic representation of the crystal structure of CeCuSn, which forms in the hexagonal GaGeLi-type structure (space group $P6_3mc$). The Ce, Cu, and Sn atoms are represented by white, gray, and black balls, respectively.

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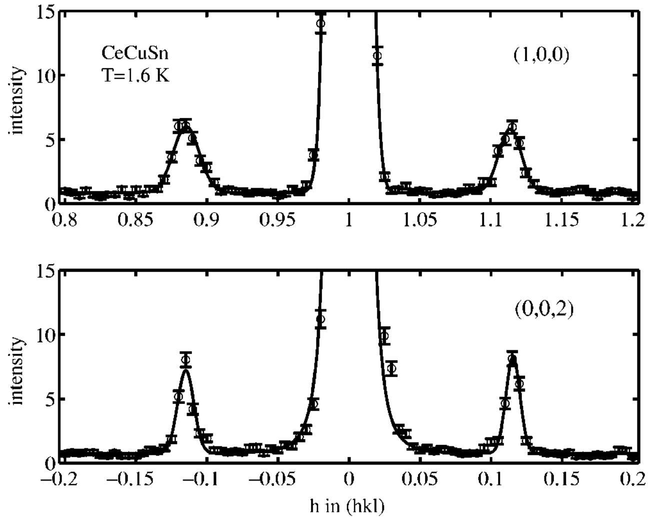

FIG. 2. Scans along the [1,0,0] direction, showing magnetic satellites near the (1, 0, 0) (upper panel) and (0, 0, 2) (lower panel) nuclear reflections.

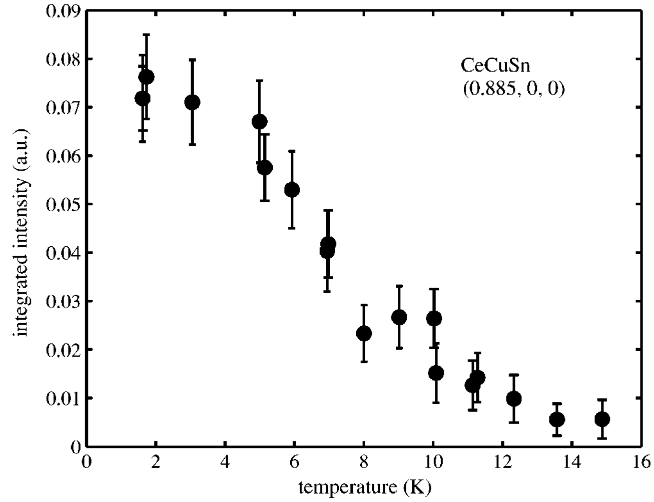

FIG. 3. Intensity of the (0.885, 0, 0) magnetic satellite as a function of temperature.

using the HB-1A triple-axis spectrometer. Incident neutrons of wavelength 2.36 Å were selected with a double pyrolitic graphite monochromator system. The sample temperature was regulated by a top-loading pumped He cryostat. In order to access a large number of reciprocal lattice points, three samples from the same batch were mounted—one with the [0, 1, 0] direction vertical, a second with the [1, 1, 0] direction vertical, and a third with the [0, 0, 1] direction vertical—allowing us to measure reflections of the types $(h,0,l)$, (h,h,l), and $(h,k,0)$, respectively, in a horizontal scattering plane. Samples were typically about 3–5 mm in diameter and 1 mm in thickness.

At 1.6 K, a series of scans along various symmetry directions revealed a set of magnetic satellites at positions $(h,k,l)\pm(0.115,0,0)$, confirming long-range antiferromagnetic order. Figure 2 shows two typical scans along the [1, 0, 0] direction; the upper panel is a scan through the reciprocal lattice point (1, 0, 0) and the lower panel is through the point (0, 0, 2). The $(1,0,0)\pm(0.115,0,0)$ reflections lie entirely in the basal plane, while the $(0,0,2)\pm(0.115,0,0)$ reflections are nearly along the c axis. The fact that both sets of satellites have intensities indicates that the magnetic moments have at least some small components in the basal plane as well as along the c axis. The observed magnetic intensities are 2-3 orders of magnitude smaller than the nuclear intensities, indicating reduced ordered moments, which may explain the difficulty in interpreting the bulk measurements. On the other hand, the magnetization (for fields applied perpendicular to the c axis) shows a tendency to saturate above about 4 T, at a value of about 1.1 μ_B/formula unit, which is a significant fraction of the full free-ion moment of Ce.

The temperature dependence of the (0.885, 0, 0) magnetic satellite is shown in Fig. 3. The intensity decreases smoothly with increasing temperature up to about 8–10 K. Above 10 K, it extends as a shallow tail up to above 12 K, and even at 15 K, one may discern a very small peak at the limits of detection. The inflection point around 8–10 K corresponds to the anomalies seen in bulk measurements. This behavior is similar to what was seen in one of the samples in Ref. 5, where the specific heat showed only a faint peak at about 10 K with a broad tail extending up to 15 K. The wave vector and peak widths (not shown here) are temperature independent within experimental uncertainties.

From single-crystal neutron diffraction experiments, we have confirmed antiferromagnetic ordering at low temperatures for CeCuSn with an incommensurate magnetic wave vector $\mathbf{k}=(0.115,0,0)$. The onset of magnetic order appears slowly around 12 K and only develops in earnest below 8–10 K. Although the magnetic ordering is completed by 1.6 K, the ordered moment is probably significantly reduced with respect to the full free-ion moment.

Recently, the isostructural compound NdCuSn was reported to order with two distinct magnetic phases in coexistence.[11] The crystal structure of NdCuSn has the same ambiguity as CeCuSn with respect to the ordering of Cu and Sn atoms. Thus, it was proposed that in NdCuSn, a majority (about 90%) of the sample orders crystallographically, and is associated with a magnetic structure with wave vector $\mathbf{q}_1=(3/8,0,0)$. The rest of the sample was believed to be crystallographically disordered and associated with a magnetic structure with wave vector $\mathbf{q}_2=(1/2,0,0)$. Based on the peak widths, the order associated with $\mathbf{q}_1$ was characterized as having rather short magnetic ordering distances. In the case of CeCuSn, μSR results have indicated the coexistence of two magnetic phases, one long-range ordered and another only short-range ordered (i.e., spin frozen).[6] Such a scenario could account for the apparently reduced ordered moment in CeCuSn and, similar to NdCuSn, could originate in crystallographic order/disorder. Preparations for more detailed structural studies to investigate this possibility are in progress.

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