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LETTER TO THE EDITOR

Crystal fields and spin dynamics of hexagonal CeTSn compounds (T = Cu, Ag and Au)

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Abstract. We report inelastic neutron scattering and magnetization measurements for hexagonal CeTSn compounds, with T = Cu, Ag and Au. Inelastic neutron spectra of all three compounds show two well defined crystal-field excitations. The overall crystal-field splitting is largest for CeAuSn. Low-energy neutron scattering studies of CeCuSn and CeAgSn reveal the presence of a quasielastic line whose width varies with the wavevector and increases weakly with temperature. Magnetization isotherms for CeCuSn at 1.7 K show several metamagnetic transitions.

The equiatomic ternary compounds CeTSn (T = Ni, Pt and Pd) crystallize in the orthorhombic TiNiSi-type structure, while those with T = Cu, Ag and Au have been reported to have the hexagonal $CaIn_2$-type structure [1–3]. Magnetic and transport measurements on these compounds reveal many unusual and interesting ground-state properties, for example the opening of a hybridization gap at low temperatures in the electronic density of states at E_F for CeNiSn [4], antiferromagnetic and Kondo lattice behaviour for CePdSn and CePtSn [5, 6], and complex antiferromagnetic ordering below 10 K in CeCuSn [7–9]. The compound CeAgSn has an antiferromagnetic ground state below 6 K, while CeAuSn remains paramagnetic down to 4.2 K [9, 10]. In order to gain more insight into the magnetic states of these compounds, we have carried out inelastic neutron scattering and magnetization measurements on the hexagonal compounds CeTSn (T = Cu, Ag and Au).

Polycrystalline samples were fabricated by arc melting under high-purity argon and were characterized using powder x-ray and neutron diffraction. This revealed that the samples were single-phase materials which crystallized in the hexagonal GaGeLi-type structure, with space group $P6_3mc$. This is a modification of the $CaIn_2$ structure in which the T and Sn sites are on ordered sublattices [11]. The cerium atoms lie on a simple hexagonal sublattice.

Inelastic neutron scattering experiments on the CeTSn compounds were performed using the HET time-of-flight spectrometer at the ISIS pulsed neutron source, Rutherford Appleton Laboratory, UK. Incident neutron energies of 35 meV and 60 meV were used, with energy resolutions of roughly 1% of these values. Phonon scattering contributions were subtracted from the observed spectra by scaling the data at high angle (118°), where the magnetic scattering is negligible. The resulting magnetic response for the CeTSn compounds is shown in figure 1. It can be seen that two very well resolved crystal-field (CF) excitations are observed in all three compounds. A superposition-model calculation for the crystal field at the Ce sites in the GaGeLi structure shows that only three parameters, B_2^0, B_4^0 and B_4^3, are required to specify the CF Hamiltonian. The positions and relative intensities of the two excitations are sufficient to fix all three CF parameters. The values of the fitted

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Table 1. Crystal-field parameters (B_n^m), ground-state magnetic moments ($\langle\mu_x\rangle$ calculated from the CF ground state, and μ_s estimated from magnetization measurements at 1.7 K) and linewidths (Γ) of the crystal-field transitions for CeTSn (T = Cu, Ag and Au) compounds.

	CeCuSn	CeAgSn	CeAuSn
B_2^0 (meV)	0.961	0.877	1.397
B_4^0 (meV)	−0.031	−0.042	−0.055
B_4^3 (meV)	0.621	0.584	1.320
$\langle\mu_x\rangle$ (μ_B/Ce)	1.19	1.19	1.12
μ_s (μ_B/Ce)	0.90	1.15 (1.2†)	—
Γ (meV)	1.97	0.93	1.16

† From reference [8], estimated from M versus H up to 35 T field at 4.2 K.

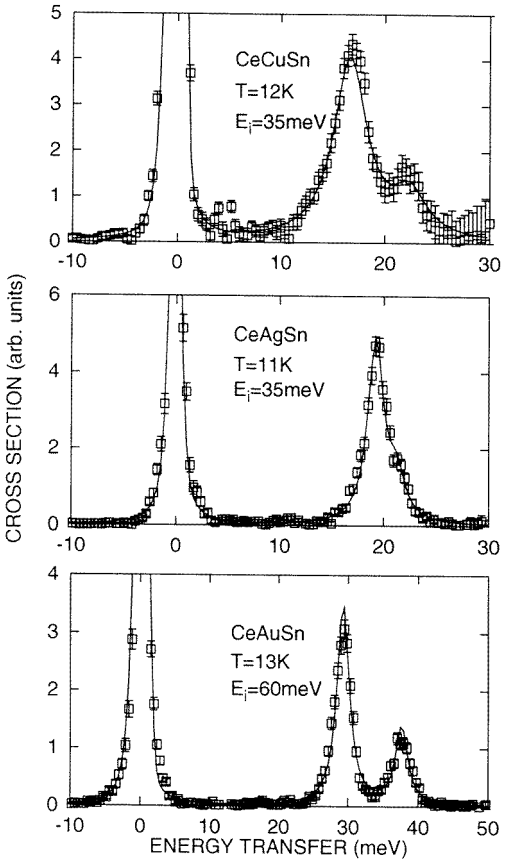

Figure 1. Magnetic scattering contributiona of the inelastic neutron spectra for CeTSn (T = Cu, Ag and Au) at low temperature, measured on the HET spectrometer. The solid lines are the fits to the crystal-field model.

CF parameters are given in table 1, and the solid lines in figure 1 are the fitted profiles for these parameters. It can be seen that the signs of the CF parameters are the same for

all three compounds, and the relative magnitudes of B_2^0/B_4^0 and B_4^0/B_4^4 are very similar, but the overall CF splittings and the CF parameters of CeAuSn are 1.5 to 2 times larger than those of CeCuSn and CeAgSn. This suggests a larger intrinsic contribution to the CF from the Au atoms. The susceptibility calculated, using the CF parameters in table 1, is strongly anisotropic with the *c*-direction a hard axis and an isotropic easy basal (*a*–*b*) plane for all three compounds. The ground-state moment calculated from the CF wavefunctions is given in table 1; this shows good agreement with that estimated from magnetization measurements.

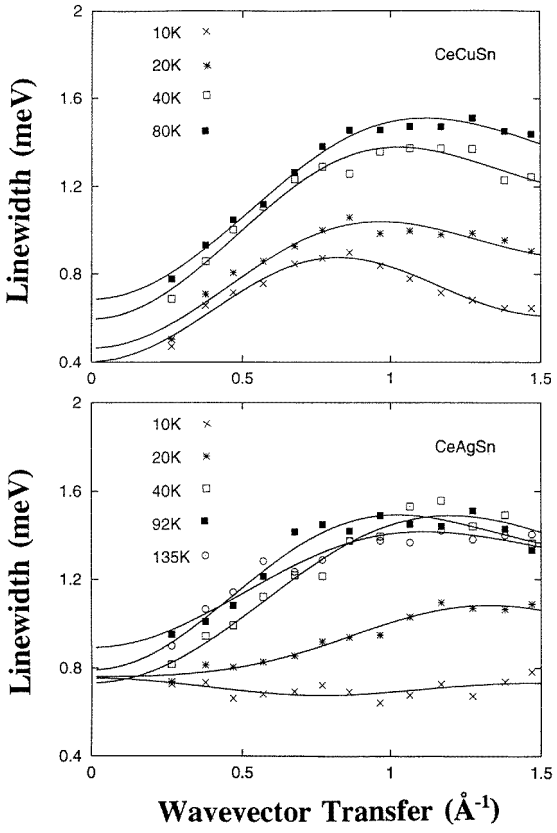

Figure 2. The quasielastic linewidth versus wavevector transfer at various temperatures for CeCuSn and CeAgSn. The solid lines are fits to a simple shell model for the RKKY interactions.

Low-energy inelastic neutron scattering was carried out using the IN6 spectrometer at the Institut Laue–Langevin, Grenoble, in order to study the temperature and wavevector dependence of the quasielastic linewidths of CeCuSn and CeAgSn. An incident neutron energy of 3.14 meV was used with an energy resolution of 0.1 meV (HWHM). For both CeCuSn and CeAgSn above T_N the magnetic response was a narrow quasielastic line with a Lorentzian line-shape. Figure 2 shows that the half-width, $\Gamma(T)$, for both compounds is modulated with the wavevector Q. The high-temperature behaviour of $\Gamma(Q, T)$ ($T \geqslant 40$ K) is what would be expected for paramagnetic scattering where the Q-modulation is due to the spin–spin coupling (RKKY interactions) between the Ce ions [12], and the linewidth is weakly temperature dependent. Below 40 K, however, the linewidth decreases almost

linearly with temperature, extrapolating to 0.5 meV at $T = 0$. This is characteristic of Kondo lattice behaviour. In CeCuSn the Q-modulation is a constant fraction of the mean linewidth, whereas in CeAgSn the modulation is strongly suppressed at low temperatures, so at 10 K the linewidth is essentially Q-independent. This is consistent with the intersite (RKKY) coupling being suppressed by the Kondo effect at low temperatures.

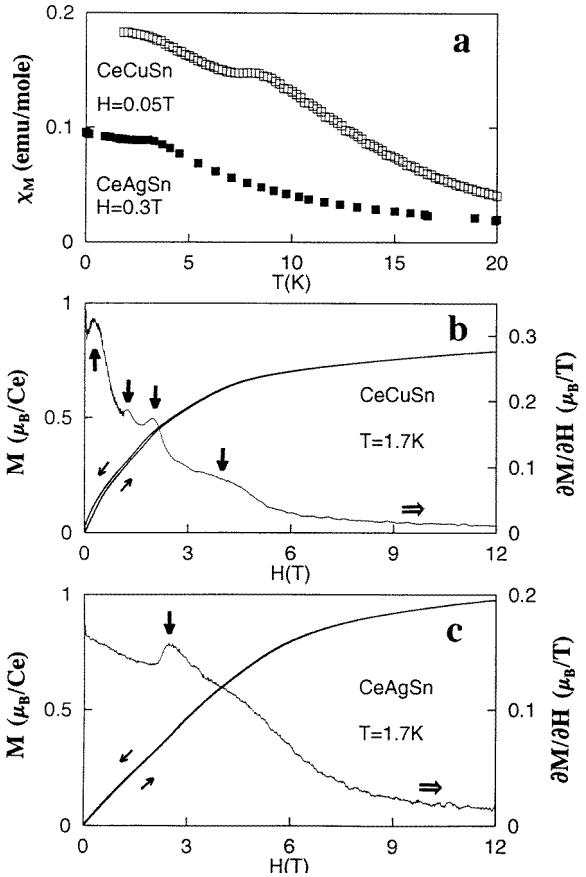

Figure 3. (a) The magnetic susceptibility versus temperature for CeCuSn and CeAgSn, (b) the magnetization and differential susceptibility (second y-axis) versus field for CeCuSn and (c) the magnetization and differential susceptibility (second y-axis) versus field for CeAgSn.

The low-temperature susceptibilities of CeCuSn and CeAgSn exhibit shoulders at 8.3 K and 6 K respectively (see figure 3(a)); this is due to antiferromagnetic ordering of the Ce moment in agreement with previous reports [7, 9, 10]. ^{119}Sn Mössbauer spectra of CeCuSn and CeAgSn at 4.2 K show a hyperfine-split pattern [10], which further confirms the magnetic ordering in these compounds. The magnetization isotherm of CeCuSn at 1.7 K exhibits several metamagnetic transitions with increasing field and a saturation-type behaviour at high fields. In order to see the metamagnetic transitions clearly, we have plotted the differential susceptibility $\partial M/\partial H$ versus H (figure 3, second y-axis) for CeCuSn and CeAgSn. $\partial M/\partial H$ for CeCuSn shows three sharp peaks and one broad peak (the positions of the peaks are marked by arrows in figures 3(b) and 3(c)), while that for CeAgSn shows a single sharp peak. This shows that the magnetic phase diagram of CeCuSn is complex. The

saturation magnetic moments were estimated by extrapolation of an M versus $1/H$ plot to $1/H \rightarrow 0$. These are given in table 1.

In conclusion, it would appear that the magnetic properties of CeCuSn, CeAgSn and CeAuSn are largely consistent with a localized Ce^{3+} state of the Ce ion. The only indication of a possible Kondo coupling is the temperature dependence of the neutron quasielastic linewidth for CeCuSn and CeAgSn below 40 K. Further signs of the Kondo effect in these compounds are (i) an anomaly in the thermopower of CeCuSn, with a positive peak at 50 K and a change of sign at 150 K [13], and (ii) the absence of magnetic ordering in CeAuSn down to 60 mK, with a γ-value of 33 mJ mol^{-1} K^{-2} [14].

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