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Journal of Magnetism and Magnetic Materials 272–276 (2004) 633–634
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Different nature of magnetism at cerium sublattices in $CeMn_2Si_2$ and $CeMn_2Ge_2$ compounds

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Abstract

The low-temperature magnetic properties of Ce atoms in the $CeMn_2Ge_2$ and $CeMn_2Si_2$ were investigated by means of (1) perturbed angular correlation experiment, and (2) ab initio density-functional calculations. They are found to be fundamentally different, despite the Ce magnetic moment is measured to be zero in both compounds. While the Ce atoms are intrinsically non-magnetic in silicide, they display a latent magnetism in the $CeMn_2Ge_2$ since their zero total moment is composed of the finite spin and orbital components which cancel each other.

PACS: 71.20.Eh; 76.80.+y; 75.20.Hr

Keywords: Rare-earth magnetism; Density-functional calculation; Perturbed angular correlation; $CeMn_2Ge_2$; $CeMn_2Si_2$

The ternary intermetallic compounds $CeMn_2X_2$ (X = Ge or Si) possess one interesting feature: they exhibit magnetic moment exclusively at Mn sites, maintaining the Ce moment quenched even at the lowest temperatures [1]. Their crystal structure is body-centered cubic (space group I4/mmm), in which the Ce atoms are surrounded octahedrally by the Mn atoms with magnetic moments aligned antiferromagnetically in the silicide, and conically (with a ferromagnetic component along the *c*-axis) in the $CeMn_2Ge_2$ (Fig. 1).

Different magnetic order of the Mn atoms in $CeMn_2X_2$ can be explained in terms of different Mn–Mn inter- and intra-layer distances [2]. Such drastic changes of the Mn magnetic configuration, however, do not seem to influence the magnetism of the Ce atoms, which exhibit a zero moment in both compounds. This surprising fact motivated us to focus attention on the Ce subsystem in these two compounds.

First, we measured a temperature dependence of the magnetic hyperfine fields (MHF) at the Ce nuclei by perturbed angular correlation (PAC) technique. The radioactive probe nucleus was ^{140}La. The 329–487 keV γ–γ cascade of ^{140}Ce, populated from β-decay of ^{140}La, has been utilized for a measurement of the magnetic interaction on its 2083 keV, 4^+ spin state. The spin–rotation spectra were recorded in the temperature range from 10 to 420 K. After the fitting, a Larmor frequency $\omega_L = g\mu_N B_{hf}/\hbar$ is deduced. Knowing the nuclear *g*-value $g = 1.12$ for the $I = 4^+$ intermediate level of ^{140}Ce, we determined the MHF B_{hf} at the Ce nuclei in the whole temperature interval, for both considered compounds.

In the case of $CeMn_2Si_2$ no finite MHF at the Ce nuclei has been registered, a result which confirmed a complete magnetic inactivity of the Ce sublattice.

In the $CeMn_2Ge_2$ case, however, the Ce nuclei exhibit a non-zero MHF, as shown in Fig. 2.

For temperatures between 120 and 320 K (Curie point) the Ce MHF follows a Brillouin function form, reflecting the ferromagnetic order of neighboring Mn moments (the so-called transferred field). If the Ce atoms were intrinsically non-magnetic, the MHF curve would follow this behavior down to $T = 0$. The

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doi:10.1016/j.jmmm.2003.12.642

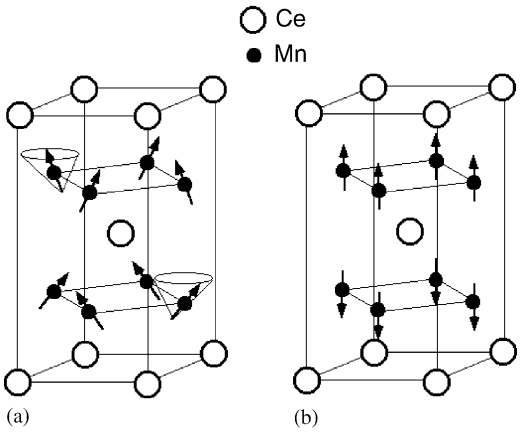

Fig. 1. Low-temperature magnetic structures of the $CeMn_2Ge_2$ (a) and $CeMn_2Si_2$ (b) compounds. The non-magnetic Ge and Si atoms are not shown.

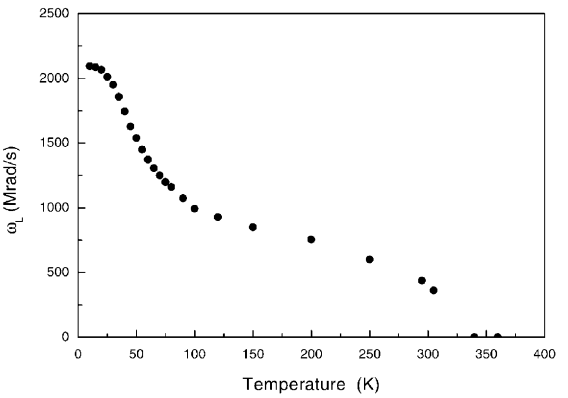

Fig. 2. Temperature dependence of the Larmor frequency measured at the Ce nuclei in the $CeMn_2Ge_2$.

low-temperature deviation of this curve, however, signalizes a magnetism of the Ce atoms, which seems to be incompatible with the zero value of the Ce magnetic moment.

In order to interpret the PAC results, we performed the first-principles calculations for both $CeMn_2X_2$ compounds using the full potential linear augmented plane wave (FP-LAPW) method, as implemented in the WIEN2k computer code. The Ce 4f states were treated as band states, and the spin–orbit coupling was taken into account.

For the $CeMn_2Si_2$ the calculations confirmed the PAC findings: the Ce atoms are found to be truly non-magnetic, with zero total magnetic moment composed of zero spin and zero orbital components.

In the case of $CeMn_2Ge_2$, the calculations gave a non-zero Ce magnetic moment of $-0.16\,\mu_B$, directed oppositely to the Mn moments and composed of finite spin ($-0.70\,\mu_B$) and orbital ($+0.54\,\mu_B$) components which almost canceled each other. Although complete cancellation did not happen (mostly due to the approximations implemented for the exchange and correlation effects), the calculations indicate that exactly this scenario occurs in the nature. Indeed, if we accept that Ce zero moment is composed of the finite spin and orbital components that completely cancel each other, then these components must create non-zero spin-dipolar and orbital contributions to MHF which, however, do not need to cancel (see formulae in Ref. [3]). The orbital MHF is much larger [4] and, when superposed to the transferred MHF, creates the low-temperature deviation of the PAC curve in the Fig. 2. It disappears at ~ 120 K, when temperature finally randomizes the Ce orbital moment.

Thus, on the basis of combined theoretical and experimental arguments we showed that magnetism of the Ce atoms has completely different nature in $CeMn_2Si_2$ and $CeMn_2Ge_2$, although in both compounds the Ce magnetic moment is measured to be zero.

References

[1] J.A. Fernandez, et al., J. Appl. Phys. 79 (1996) 5398.
[2] J.H.V.J. Brabers, et al., Phys. Rev. B 50 (1994) 16410.
[3] S. Blügel, et al., Phys. Rev. B 35 (1987) 3271.
[4] M.V. Lalić, et al., Phys. Rev. B 65 (2002) 054405.