Magnetic-field dependence of spin correlation in chiral lattice semimetal $Ce_3Co_4Sn_{13}$

K. Iwasa[1], Y. Otomo[2], and J.-M. Mignot[3]
Frontier Research Center for Applied Atomic Sciences, Ibaraki University, Japan[1]
Department of Physics, Tohoku University, Japan[2]
Laboratoire Léon Brillouin, CEA-CNRS, CEA/Saclay, France[3]

It is an attractive issue to investigate electronic correlation phenomena associated with topological symmetries. For example, massless Dirac/Weyl fermions have been extensively studied, which are characterized by linear dispersion for electron bands and have been identified for the two-dimensional systems. Recently, Dirac/Weyl electrons are also searched in three-dimensional (3D) bulk systems. It was discussed in a theoretical study that such 3D Weyl fermions intrinsically appear in chiral symmetry lattices [J. L. Manñs, Phys. Rev. B **85**, 155118 (2012)].

In the case of f-electron systems based on rare-earth alloy compounds, the c–f effect has been a key issue. Kondo semimetals or semiconductors exhibit band gap features as a consequence of such a hybridization effect. We expect that the Weyl fermions are formed in a Kondo semimetal taking a chiral symmetry structure. Considering such electronic state, we have investigated a class of $Ce_3Tr_4Sn_{13}$ (Tr: transition-metal elements).

$Ce_3Co_4Sn_{13}$ undergoes a structural phase transition at 160 K [C. S. Lue et al., Phys. Rev. B **85**, 205120 (2012)]. We evidenced that the low-temperature crystal structure takes the chiral space group $I2_13$ [Y. Otomo et al., Phys. Rev. B **94**, 075109 (2016)]. The electrical resistivity data of $Ce_3Co_4Sn_{13}$ is less dependent on temperature compared to metallic behaviors of $La_3Co_4Sn_{13}$ without $4f$ electrons. These facts are expected to indicate the formation of Weyl semimetal state in $Ce_3Co_4Sn_{13}$. The electronic Sommerfeld coefficients of these compounds was evaluated to be approximately 4 J/(mol-Ce K^2) at 1 K [A. L. Cornelius et al., Physica B **378–380**, 113 (2006), A. Ślebarski et al., Phys. Rev. B **86**, 205113 (2012), E. L. Thomas et al., J. Solid State Chem. **179**, 1642 (2006)]. This fact was understood as HF systems. However, our recent inelastic neutron scattering (INS) experiment revealed the emergence of spin excitations in the range up to 1 meV below 20 K [K. Iwasa et al., Phys. Rev. B **95**, 195156 (2017)]. On the other hand, $Ce_3Co_4Sn_{13}$ does not exhibit any magnetic ordering down to 0.5 K, and previous study on $Ce_3Co_4Sn_{13}$ reported the field-induced antiferromagnetic correlations at 4.2 K [A. D. Christianson et al., Physica B **403**, 909 (2008)]. Based on the $I2_13$ structure, the two inequivalent Wyckoff sites for the Ce ions take different CEF schemes [K. Iwasa et al., Phys. Rev. B **95**, 195156 (2017)]. Thus, the magnetic-field-induced antiferromagnetic correlation can be understood different magnetic moments at the two Ce-ion sites. In order to obtain microscopic information for the Ce $4f$-electron state, we performed INS measurements under the magnetic fields up to 6 T below 20 K, by using the cold-neutron spectrometer 4F2 installed in the Orphée reactor of Laboratoire Léon Brillouin.

Upper part of Fig. 1 shows INS spectra at the scattering vector $\mathbf{Q} = (1, 0, 0)$ for the original high-temperature unit cell of $Ce_3Co_4Sn_{13}$ measured at 1.6 and 20 K under magnetic fields of zero and 4 T applied along the $[0, -1, 1]$ axis. The data at 1.6 K and 4 T show a slight intensity enhancement near the excitation energy of 0.25 meV. The data measured at $\mathbf{Q} = (1.25, 0, 0)$ also show an increase in the inelastic-scattering intensity, although

the result is not shown here. In addition, we observed a drastic increase in intensity in the elastic scattering region of $\mathbf{Q} = (1, 0, 0)$, as shown by open marks of Fig. 1. Asymmetric spectral shape is considered to be due to alignment of four single-crystal samples. This intensity enhancement occurs below approximately 10 K. In contrast, such elastic-intensity enhancement was not observed at $\mathbf{Q} = (1.25,\ 0,\ 0)$, $(1.5,\ 0,\ 0)$, and $(1,\ 1,\ 1)$, where the signals of collective spin excitation were observed in previous zero-field INS measurements. Lower part of Fig. 1 shows a temperature dependence of the elastic-scattering intensity at $\mathbf{Q} = (1,\ 0,\ 0)$. The intensity at 1.6 K (red circles) shows a convex curve of magnetic fields, and is saturated above approximately 5 T. In contrast, the intensity at 5 K (green squares) follows a function of the squared magnetic field, as shown by a solid line fitted to the data. The data at 20 K (blue diamonds) exhibit no magnetic-field dependence. The data at 5 K indicate that the magnitude of field-induced magnetic moment shows a linear relationship to the magnetic fields, which corresponds to a paramagnetic behavior. However, the convex behavior of the data at 1.6 K is in marked contrast to the paramagnetic-like behavior. This phenomenon is rather close to that of a ferromagnetic spin correlation.

The spin correlation of the $4f$ electrons in $Ce_3Co_4Sn_{13}$, which is characterized by the signal near $\mathbf{Q} = (1,\ 0,\ 0)$, is enhanced by applied magnetic fields below 10 K. According to the study on electrical resistivity [J. R. Collave et al., J. Appl. Phys. **117**, 17E307 (2015)], the resistivity is suppressed by applied magnetic fields. This phenomenon indicates that the magnetic-field induced increase in carrier number causes a stronger RKKY-type interaction. Therefore, we expect magnetic-field tuning of the quantum criticality in the chiral-lattice symmetry of $Ce_3Co_4Sn_{13}$.

We thank P. Boutrouille for technical support on performing the neutron scattering measurements. The experiments were supported by General User Program for Neutron Scattering Experiments, Institute for Solid State Physics, The University of Tokyo, at JRR-3, Japan Atomic Energy Agency, Tokai, Japan.

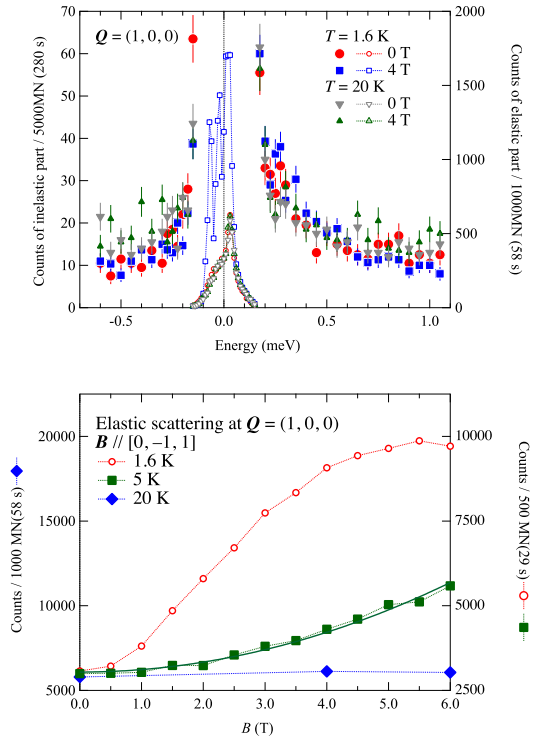

Fig. 1. (Upper part) INS spectra at $\mathbf{Q} = (1, 0, 0)$ measured at several temperatures and magnetic fields. (Lower part) Magnetic-field dependences of elastic-scattering intensity at $\mathbf{Q} = (1,\ 0,\ 0)$.