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Journal of Alloys and Compounds 451 (2008) 504–506

Journal of ALLOYS AND COMPOUNDS

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Ground state magnetic structure of $Ce_2Ni_3Ge_5$

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Available online 19 April 2007

Abstract

We have carried out neutron diffraction experiments on $Ce_2Ni_3Ge_5$ single crystal. $Ce_2Ni_3Ge_5$ orders antiferromagnetically below $T_N = 5$ K and exhibits a successive transition at $T^* = 4.3$ K. We have confirmed that the ground state antiferromagnetic structure is characterized with the propagation vector $q = [0\,1\,0]$. We have revealed that this magnetic structure stabilizes only below $T^* = 4.3$ K from the simultaneous neutron diffraction and electrical resistivity measurements, contrary to the previously reported neutron powder diffraction result. From the analysis of magnetic reflection intensities, we suggest that the ground state magnetic structure is a canted AF structure, where the moments orient about 20° tilted from the *a*-axis in the *ab*-plane. The obtained AF components are 0.45 μ_B/Ce along the *a*-axis and 0.15 μ_B/Ce along the *b*-axis. It is suggested that the high temperature phase is characterized with another magnetic phase having a different propagation vector, which is not determined in the present study.

Keywords: $Ce_2Ni_3Ge_5$; Neutron diffraction; Antiferromagnet

1. Introduction

The study of electronic structures in ternary Ce–Ni–Ge compounds has been receiving increasing interest after the discovery of superconductivity at ambient and under high pressure in a non-Fermi-liquid compound $CeNi_2Ge_2$ [1]. Recently, it is reported that a pressure-induced superconductivity (PIS) is realized in $CeNiGe_3$ and $Ce_2Ni_3Ge_5$ around the quantum critical point (QCP), where the magnetic ordering temperature vanishes [2,3]. It is quite important to investigate magnetic properties in these compounds at ambient and under high pressure in order to clarify the mechanism of PIS at QCP.

$Ce_2Ni_3Ge_5$ which crystallizes in the $U_2Co_3Si_5$-type orthorhombic structure (Ibam) is an antiferromagnet (AFM) with $T_N = 5.0$ K. Magnetization and specific heat measurements revealed the successive anomalies at T_N and $T^* = 4.3$ K [4,5]. From the specific heat measurement, it is suggested that the transitions at T_N and T^* are the second and first-order transitions, respectively. With increasing pressure, the AFM is suppressed and a superconducting ground state is induced below 0.26 K around the critical pressure of 3.9 GPa [3].

The AFM structure in $Ce_2Ni_3Ge_5$ is searched by a neutron powder diffraction experiment and determined as: (i) the AFM structure is collinear with a propagation vector $q = [0\,1\,0]$, where a ferromagnetic *b*-plane stacked antiferromagnetically along the *b*-axis with "$++--$" or "$+--+$" manner, (ii) the magnetic moment which is parallel to the *a*-axis is about 0.4 μ_B/Ce at 1.4 K and (iii) the AFM structure stabilizes below $T_N = 5$ K [6]. While it is claimed that magnetic easy axis is most likely along the *c*-axis and moment size on Ce is expected to be higher from the recent magnetic susceptibility measurement and its analysis with CEF model [5]. In order to clarify the magnetic structures of $Ce_2Ni_3Ge_5$, neutron diffraction experiments on a single crystal have been carried out.

2. Experimental

A single crystal of $Ce_2Ni_3Ge_5$ with the size of 1 mm × 1 mm × 2 mm has been grown by the Bi-flux method. The detailed description for the sample preparation is reported elsewhere [5]. Neutron diffraction experiments have been carried out using the thermal and cold triple-axis spectrometers TAS-2 and LTAS, and the newly constructed double-axis spectrometer MUSASI installed at the research reactor JRR-3 in the Japan Atomic Energy Agency, JAEA. In order

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0925-8388/$ – see front matter © 2007 Published by Elsevier B.V.
doi:10.1016/j.jallcom.2007.04.191

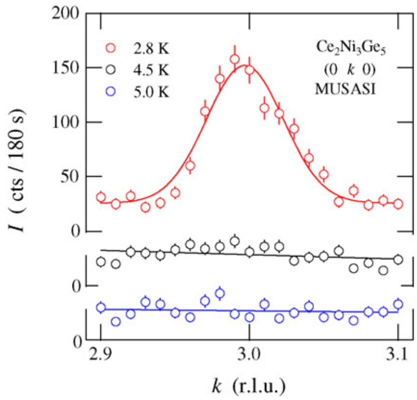

Fig. 1. Line scan around (0 3 0) in different thermodynamic phases at 2.8, 4.5 and 5.0 K.

to clarify the magnetic order parameter around the magnetic transition temperatures, we have carried out simultaneous measurements of neutron diffraction and electrical resistivity.

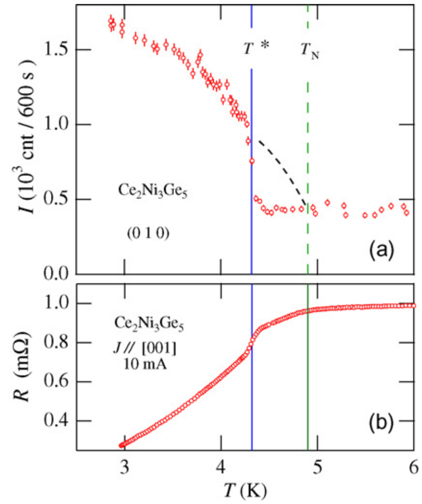

Fig. 2. Temperature dependence of: (a) magnetic reflection intensity at (0 1 0) and (b) electrical resistivity along the c-axis in $Ce_2Ni_3Ge_5$. The dashed line is the extrapolated magnetic order parameter towards $T > T^*$.

3. Results and discussions

Fig. 1 shows the respective line scans around (0 3 0) in the low temperature ($T = 2.8$ K), high temperature (4.5 K) ordered phases and paramagnetic phase (5.0 K). At the lowest temperature, we observed the antiferromagnetic super-lattice reflection with the propagation vector $q = [0\,1\,0]$. This propagation vector is consistent with the previously reported neutron powder diffraction result. It should be noted that the magnetic reflection disappears at 4.5 K, although it was claimed that this magnetic propagation stabilized below $T_N = 5$ K in the previous study [6].

In order to elucidate the stability of the ground state magnetic phase of $Ce_2Ni_3Ge_5$, we measured temperature dependence of (0 1 0) magnetic reflection and electrical resistance along the c-axis simultaneously. The magnetic intensity disappears at T^* with increasing temperature, indicating that the magnetic structure stabilizes *only* below T^*. The order parameter shows the first-order like transition, which is consistent with the specific heat anomaly [5]. The dashed line in Fig. 2 is the extrapolation of the order parameter towards $T > T^*$ region. It seems that the magnetic order parameter changes continuously up to T_N. This implies that the magnetic moment changes continuously to the high temperature phase which can be characterized with another magnetic phase having a different propagation vector. In other words, there can be two magnetically ordered phases below T_N in $Ce_2Ni_3Ge_5$, and the magnetic order parameter in the high temperature phase would behave like the dashed line. Such a continuous change of the magnetic order parameter at the magnetic order–order transition is also seen in UCu_2Si_2 [8].

The obtained (◯, left axis) and calculated (bars, right axis) integrated intensity of magnetic reflections of $Ce_2Ni_3Ge_5$ at 2.8 K is displayed in Fig. 3. We assumed two antiferromagnetic components in the present calculation, which are Γ_{3a} and Γ_{3b} of Bertaut's representation [6,7], as shown in the inset. Γ_{3a} and Γ_{3b} correspond to the AFM components along the a- and b-axis, respectively. It is noted that Γ_{3a} is one of the proposed magnetic structures for $Ce_2Ni_3Ge_5$ in ref. [6], where ferromagnetic b-planes stack along the b-axis with "+ − − +" manner. Several reflections, such as (0 1 2), (0 0 1) and (3 2 0), are not expected by the Γ_{3a} type of AF structure. Those reflections are well reproduced assuming the b-axis component, which is Γ_{3b}, where the moments stack along the b- and c-axes with "+ − + −" manner. In the calculation, we assumed 0.45 μ_B/Ce and 0.15 μ_B/Ce along the a-axis and b-axis, respectively. The estimated total magnetic moment is about 0.5 μ_B/Ce at 2.8 K and is tilted about 20° from the a-axis in the ab-plane.

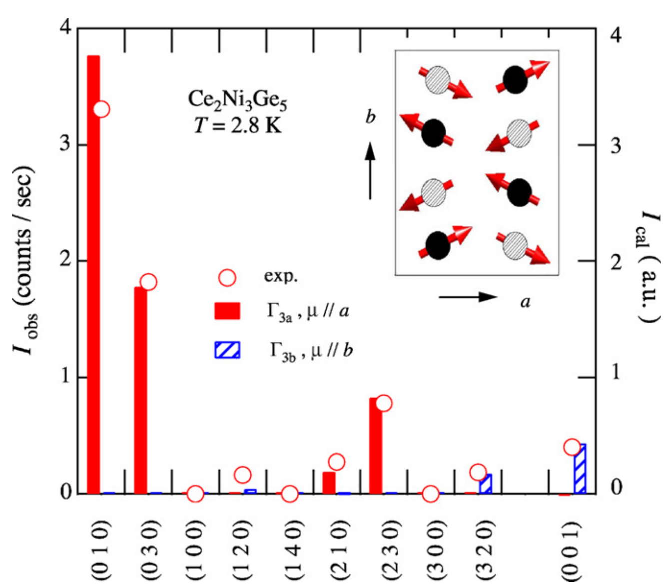

Fig. 3. The observed integrated intensity of the antiferromagnetic reflections of $Ce_2Ni_3Ge_5$ (◯, left axis), compared with model calculations (right axis), where the antiferromagnetic moments are parallel to the a-axis (solid bars) and b-axis (hatched bars). The assumed magnetic structures projected onto the (0 0 1) plane are shown in the inset. Ce atoms are indicated by black circles for $z = 0$ or 1, and hatched circles for $z = 1/2$.

The proposed ground state magnetic structure of $Ce_2Ni_3Ge_5$ observed in the present high quality single crystal is somehow related to but different from the one determined in the previous study [6]. A similar canted AFM structure was reported for iso-structural ternary silicide $U_2Rh_3Si_5$ [9]. In order to clarify the proposed canted structure in $Ce_2Ni_3Ge_5$, a spin polarized neutron diffraction experiment is in progress.

Magnetic structure of the high temperature ordered phase is still unknown. There are several compounds exhibiting two magnetic transitions in the $R_2T_3X_5$ system [10,11], while the magnetic structures of these compounds are rarely investigated. We have searched magnetic peaks at 4.5 K in several scattering planes, e.g. ($h\,k\,0$), ($h\,k\,0$) and ($h\,0\,l$), but we have not found any of them so far. Further neutron diffraction experiments are desirable for determining the high temperature magnetic structure.

Acknowledgment

A part of the present study was financially supported by the Grant-in-Aid for Scientific Research from the Japanese Ministry of Education, Culture, Sports, Science and Technology (No. 18740219).

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