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SPECIFIC HEAT AND MAGNETIC STRUCTURE OF GdT_2X_2 COMPOUNDS (T = Cu, Ni, X = Sn, Sb)

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The magnetic specific heat of $GdNi_2Sn_2$, $GdNi_2Sb_2$ and $GdCu_2Sb_2$ shows a λ-type anomaly at T_N with a noticeable reduction of the discontinuity compared to that expected for a simple antiferromagnet. It can be associated with the existence of amplitude modulated magnetic structure just below T_N. Above T_N a weak magnetic contribution is observed, evidencing the short-range ordering. From neutron diffraction studies it can be also concluded that investigated compounds have not simple antiferromagnetic structures. For $GdNi_2Sn_2$ the propagation vector $\boldsymbol{Q} = (0.3, 1/2, 1/2)$ was found with the magnetic moments along the [1,0,0] axis.

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1. Introduction

The ternary intermetallic RT_2X_2 compounds (R = rare earth, T = transition metal and X = an element of the IV or V group) exhibit a great diversity of magnetic properties [1]. In particular, many compounds present long period or incommensurate magnetic structures at low temperatures. These properties are determined by the rare earth moments, by their exchange interactions mediated by conduction electrons (RKKY type) and by effects of crystalline electric fields (CEF) acting on the $4f$ electrons. Studies of compounds with gadolinium are interesting since CEF effects can be neglected.

The ternary stannides and antimonides crystallize with the primitive tetragonal $CaBe_2Ge_2$-type of structure with space group $P4/nmm$. Intermetallic compounds $GdNi_2Sn_2$, $GdNi_2Sb_2$ and $GdCu_2Sb_2$ order antiferromagnetically below 7 K, 6 K and 11 K, and magnetic interactions seem to be frustrated. In ordered range anomalies in magnetization vs. magnetic field curves, in resistivity and in susceptibility may be attributed to the changes of magnetic structure. Below T_N the $\rho(T)$ dependences differ from that calculated for a collinear antiferromagnetic system with equal Gd magnetic moments [2]. These experimental data are indicative of a noncollinear or amplitude-modulated magnetic ordering just below T_N. In order to elucidate these phenomena the compounds were studied by neutron diffraction and specific heat measurement.

2. Neutron diffraction

The huge absorption cross-section of natural gadolinium at thermal neutron wavelengths creates severe problems for neutron diffraction studies in Gd based compounds. This is a reason why magnetic structures were solved for three GdT_2X_2 compounds only: $GdCu_2Si_2$ [3], $GdNi_2Si_2$ [3] and $GdPt_2Si_2$ [4].

Neutron diffraction studies of polycrystalline samples $GdNi_2Sn_2$, $GdNi_2Sb_2$ and $GdCu_2Sb_2$ were performed on the 7C2 diffractometer in the L. Brillouin Laboratory in Saclay using the wavelength $\lambda = 0.56$ Å. The magnetic reflections observed at low temperature (Fig. 1) cannot be indexed in a simple way, therefore

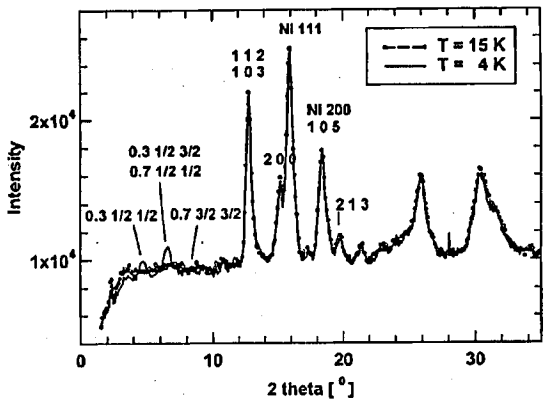

Fig. 1. Neutron diffraction patterns for $GdNi_2Sn_2$ at 15 K and 4 K.

it can be concluded that investigated compounds have not simple antiferromagnetic structures. Because of the poor resolution of the diffractometer for that wavelength, it is very difficult to find propagation vectors. Only for $GdNi_2Sn_2$ the propagation vector $Q = (0.3, 1/2, 1/2)$ was found with the magnetic moments along the [1,0,0] axis. However, it should be mentioned that the small resolution of these patterns does not allow to choose between noncollinear structure (with equal moments) and collinear sine-wave modulated structure.

3. Specific heat

Specific-heat measurements were carried out by using ac calorimetry technique [5], at temperature ranging from 1.7 K to 40 K. Magnetic contributions to the specific heat in three investigated compounds are shown in Fig. 2. It is obtained by subtracting of the lattice contribution from the rough data. The specific heat of the isomorphous nonmagnetic compound $LaNi_2Sb_2$ [6] was taken as the lattice contribution in $GdNi_2Sb_2$. The lattice contributions for $GdNi_2Sn_2$ and $GdCu_2Sb_2$ compounds were approximated by Debye functions with $\Theta_D = 225$ K and $\Theta_D = 200$ K, respectively. The shape of $C_{mag}(T)$ dependence is slightly different for three investigated compounds: a well-defined λ-type anomaly for $GdNi_2Sn_2$, a negative curvature below T_N for $GdNi_2Sb_2$ and the widest λ-type anomaly for

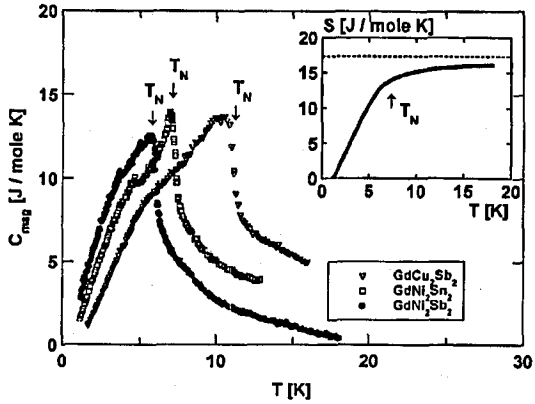

Fig. 2. Magnetic contribution to the specific heat. Inset: magnetic entropy of $GdNi_2Sb_2$; the dashed line indicates the $R\ln 8$ limit.

$GdCu_2Sb_2$. Above T_N there is a short-range order contribution. A way to evaluate the importance of these short-range correlations is to consider a magnetic entropy S. For example in the inset of Fig. 2 the magnetic entropy of $GdNi_2Sb_2$ is shown. It reaches $S_m = 16.2$ J/(K mole) at $T = 3T_N$. Estimating the missing contribution below 1.6 K as about 1 J/(K mole) leads to the total entropy $S_m = 17.2$ J/(K mole), a value very close to the theoretical value for 1 mole of Gd^{3+} ions, i.e. $R\ln 8 = 17.3$ J/(K mole). Thus the relative proportion of magnetic entropy above Néel temperature with respect to the full value is about 15%, this value is typical of many Gd compounds [5]. Just below T_N the magnetic specific heat attains a maximum value of 14.5 J/(K mole) for $GdCu_2Sb_2$, 14.0 J/(K mole) for $GdNi_2Sn_2$ and only 12.5 J/(K mole) for $GdNi_2Sb_2$, values noticeably smaller than that expected for an equal moments structure i.e. $\Delta C = 20.2$ J/(K mole). Such important reductions of the λ-type anomaly were observed for different Gd compounds [4, 5], they are consistent with the value of the $\Delta C = 13.4$ J/(K mole) predicted for an amplitude modulated structure just below T_N [7]. The additional anomalies in the $C_{mag}(T)$ dependence are seen below T_N at temperatures corresponding to anomalies in the $\rho(T)$ and $\chi(T)$ dependences [2], which are attributed to a change of the magnetic structure.

4. Conclusions

From resistivity, susceptibility, specific heat and neutron diffraction results one may reasonably conclude that none among the three investigated compounds exhibits a simple antiferromagnetic structure. As a consequence of oscillating RKKY-type exchange interactions in these compounds, an amplitude modulated structure is likely stabilized just below an ordering temperature. Moreover, as the temperature is decreased, the amplitude modulated structure must evolve towards equal moments structure in line with anomalies observed in resistivity, specific heat and susceptibility.

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