Magnetic Properties of Geometrical Frustration System : Tb$Pd_{1-x}Ni_x$Al

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Abstract

Magnetic Tb ions of the ternary rare-earth intermetallic compounds TbXAl (X = Pd and Ni) with the hexagonal ZrNiAl-type structure form the kagomé lattice in the basal plane. In order to study substitutional effect on magnetic frustration in these system, we have carried out experiments of X-ray diffraction and magnetic susceptibility in Tb$Pd_{1-x}Ni_x$Al polycrystalline samples. The x-dependence of the lattice constant c has a maximum at $x = 0.4$. On the other hand, the x-dependences of the Néel temperature T_{N1} and the paramagnetic Curie temperature θ_p show a minimum at $x \approx 0.4$, respectively. These results suggest substitutional effects on Tb$Pd_{1-x}Ni_x$Al are mainly governed by the exchange interaction along the c-axis.

Key words: magnetic susceptibility; frustration; Tb$Pd_{1-x}Ni_x$Al

The ternary rare-earth intermetallic compounds TbXAl (X = Pd and Ni) crystallize in the hexagonal ZrNiAl-type structure, where the hexagonal phase of TbPdAl is metastable above about 1,000 °C. Magnetic Tb ions are arranged in the basal plane with a triangular coordination symmetry similar to the kagomé lattice. Two of those layers are separated by a nonmagnetic layer, containing X and Al atoms. Both compounds exhibit common magnetic properties as follows [1–3]: (1) the existence of antiferromagnetically successive phase transition temperatures T_{N1} and T_{N2} (T_{N1} = 43 K and T_{N2} = 22 K in TbPdAl and T_{N1}= 47K and T_{N2} = 23 K in TbNiAl), (2) an Ising-like antiferromagnetic ordering due to a strong magnetocrystalline anisotropy, (3) reduction of the amplitude of the ordered moment at some crystallographically equivalent sites in the intermediate temperature region.

Their anomalous magnetic properties in TbXAl have been discussed in term of a geometrical magnetic frustration. Antiferromagnetic exchange coupling between Tb ions together with the strong magnetocrystalline anisotropy gives rise to the formation of geometrical frustration of magnetic moments. Since the crystal structure of both ternary compounds is identical and substituting Ni for Pd will not change the number of free electrons crucially, one can expect a chemical pressure effect and a random effect on the geometrical magnetic frustration.

In order to investigate substitutional effect on the magnetic frustration system TbXAl, we have performed experiments of X-ray diffraction and magnetic susceptibility in Tb$Pd_{1-x}Ni_x$Al polycrystalline samples. In this paper, we present strong correlation between magnetic characteristic temperatures and the lattice constant c in pseudoternary compounds Tb$Pd_{1-x}Ni_x$Al.

Polycrystalline samples of Tb$Pd_{1-x}Ni_x$Al were synthesized by arc-melting stoichiometric mixtures of pure elements (Tb: 3N, Pd: 3N5, Ni:4N and Al: 5N) in a pure Ar atmosphere. The X-ray powder diffraction pat-

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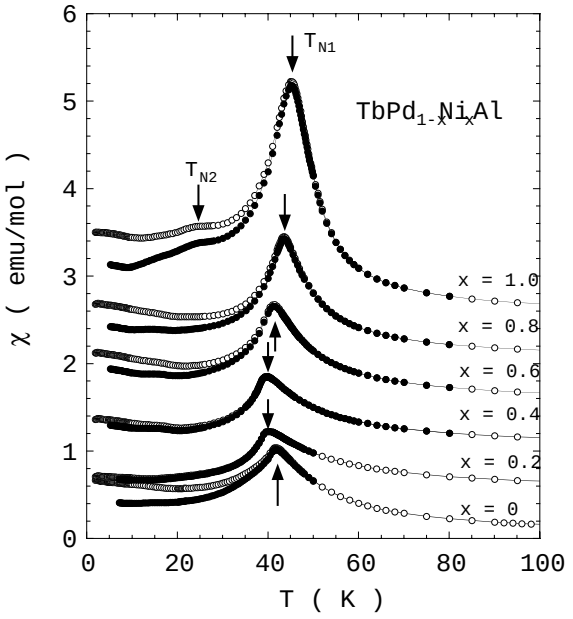

Fig. 1. Temperature dependence of the magnetic susceptibility in Tb$Pd_{1-x}Ni_x$Al. The data were measured for 0.1 T after cooling the sample from 80 K to 1.9 K at zero field by solid circles (χ(ZFC)) or in the field of 0.1 T by open circles (χ(FC)). Each curve is shifted by +0.5 emu/mol along the vertical axis.

terns at room temperature for as-cast samples could be indexed completely with the hexagonal ZrNiAl-type structure. The lattice constants were calculated by the RIETAN Rietverd analysis program.[4] The DC magnetic susceptibility from 1.9 to 300 K under 0.1 T was carried out by using a SQUID magnetometer (Quantum Design Ltd.).

Figure 1 shows the temperature dependence of magnetic susceptibility $\chi(T)$ for Tb$Pd_{1-x}Ni_x$Al. The Néel temperature T_{N1} was defined by the temperature where $\chi(T)$ displays a maximum. A small hump at 25 K in $\chi(T)$ curves for $x = 1$ should be attributed to magnetic ordering at T_{N2}. However, it was difficult to assign some anomaly to determination of T_{N2} from $\chi(T)$ curves for $x \neq 1$ below T_{N1}. The Curie-Weiss temperature θ_p was determined from fitting to the Curie-Weiss law $\chi(T) = C/(T - \theta_p)$ in the paramagnetic region, where C is the Curie constant. The difference of $\chi(T)$ between the zero-field-cooled (ZFC) and field- cooled (FC) processes decreases with increase of x, disappears at $x \approx 0.4$ and increases again to $x = 1$. The difference of $\chi(T)$ may be related to frustration effects of the magnetic moments.

The amplitude of T_{N1} and θ_p determined from $\chi(T)$ curves is plotted as a function of x in Fig. 2. The positive θ_p and antiferromagnetic ordering indicate strong competition between antiferromagnetic and ferromagnetic interactions. Both T_{N1} and θ_p display minima at $x \approx 0.4$. If one considers the random effect on mag-

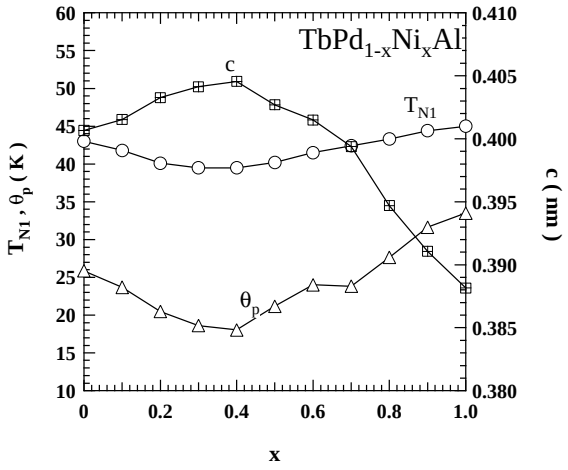

Fig. 2. Characteristic temperatures and lattice constant c as a function of Ni concentration x in Tb$Pd_{1-x}Ni_x$Al.

netism of Tb$Pd_{1-x}Ni_x$Al, those minima should appear at $x \approx 0.5$. When one compares those x-dependences of T_{N1} and θ_p with those of lattice constants, only the lattice constant c seems to have relation to T_{N1} and θ_p as shown in Fig.2. However, the lattice constant a shows a minimum at $x \approx 0.8$ and an average lattice constant $(\sqrt{3}a^2c/2)^{1/3}$ decreases monotonically from $x = 0$ to x =1. The above-mentioned coincidence suggests that magnetism in Tb$Pd_{1-x}Ni_x$Al is more sensitive to change of interactions along the c-axis than the random effect and change of interactions in the basal plane. Each Pd or Ni atom is located at a center of a ring consisting of six Tb-triangles (1a site) or a body-center of a trigonal prism with six corners of Tb atoms (2d site). The substitution at 1a site will not strongly affect the frustration in kagomé lattice, while the substitution at 2d site will have great influence on the magnetic coupling between basal planes.

To conclude, we have carried out experiments of X-ray diffraction and magnetic susceptibility in the geometrical frustration system Tb$Pd_{1-x}Ni_x$Al. The x - dependence of T_{N1}, θ_p and c indicates that substitutional effects on Tb$Pd_{1-x}Ni_x$Al are mainly governed by the exchange interaction along the c-axis.

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