14th Czech and Slovak Conference on Magnetism, Košice, Slovakia, July 6–9, 2010

Magnetic Properties and Specific Heat of Laves Phase $Tb_{1-x}Sc_xNi_2$ ($x = 0.1$, 0.2) Solid Solutions

J. Ćwik[a,*], T. Palewski[a], K. Nenkov[a,b], J. Lyubina[b], J. Warchulska[a], O. Gutfleisch[b] and J. Klamut[a,c]

[a]International Laboratory of High Magnetic Fields and Low Temperatures, Gajowicka 95, 53-421 Wrocław, Poland

[b]IFW Dresden, Postfach 270016, 01171 Dresden, Germany

[c]Institute of Low Temperature and Structure Research, PAS, Okólna 2, 50-422 Wrocław, Poland

Magnetic and specific heat measurements have been performed on polycrystalline $TbNi_2$, $ScNi_2$ and their solid solutions $Tb_{1-x}Sc_xNi_2$ ($x = 0.1$, 0.2). These compounds were synthesized using high-purity rare-earth metals. It has been found that the magnetic susceptibility of the nonmagnetic $ScNi_2$ compound exhibits a very weak temperature dependence characteristic of the Pauli paramagnets. $TbNi_2$, $Tb_{0.9}Sc_{0.1}Ni_2$ and $Tb_{0.8}Sc_{0.2}Ni_2$ are typical Curie–Weiss paramagnets and are ferromagnetically ordered below 36 K. As revealed by room-temperature X-ray powder diffraction all the $Tb_{1-x}Sc_xNi_2$ solid solutions have the cubic Laves $C15$-type superstructure. The Debye temperature, phonon and conduction electron contributions as well as the magnetic part of heat capacity were determined. The magnetocaloric effect has been studied by means of specific heat measurements in magnetic fields of 0.42 and 1 T. The effect of rare-earth substitution in $ScNi_2$ on the magnetic and magnetocaloric properties will be discussed.

PACS numbers: 75.30.−m, 75.30.Cr, 75.40.−s, 75.85.+t

1. Introduction

The magnetism properties of compounds with exchange interaction comparable in magnitude to the crystal field splitting are of particular fundamental interest. In the solid solutions $Tb_{1-x}Sc_xNi_2$, the substitution of Tb ions having nonzero magnetic moment for nonmagnetic scandium ions leads to the suppression of the long-range magnetic order of Tb ions. The RNi_2 intermetallic compounds were the subject of extensive investigations in the past decades because of their relatively simple structure and the commonly accepted assumption that Ni carries no magnetic moment [1–3]. $TbNi_2$ is a typical Curie–Weiss paramagnet at $T > T_C$ and is ferromagnetically ordered at a relatively low temperature. According to the latest information the Curie temperature of $TbNi_2$ is 36 K [4]. The $ScNi_2$ compound exhibits only the Pauli paramagnetism in the temperature range of 2–298 K [5]. The purpose of this work is to study and analyse physical properties including the magnetocaloric effect (MCE) of $Tb_{1-x}Sc_xNi_2$ solid solutions upon substitution of Tb by Sc in the magnetically ordered $TbNi_2$, as well as those in the nonmagnetic $ScNi_2$ compound.

2. Experimental

The methods for sample preparation and quality control were described in [6]. Specific heat measurements were performed in the temperature range of 1.9–295 K in applied magnetic fields of 0.42 and 1 T using Quantum Design PPMS 14 Heat Capacity System. Temperature dependence of the DC magnetic susceptibility were measured at 4.2–290 K in a static magnetic field of 0.42 T using a Faraday balance method. X-ray powder diffraction revealed that $Tb_{1-x}Sc_xNi_2$ samples are single phase and have the cubic $C15$ superstructure (space group $F\bar{4}3m$). The lattice parameters of $TbNi_2$, $Tb_{0.9}Sc_{0.1}Ni_2$ and $Tb_{0.8}Sc_{0.2}Ni_2$ were determined to be 1.432, 1.428, and 1.423 nm, respectively.

3. Results and discussions

Figure 1 shows the dependence of the magnetic susceptibility for all measured samples in the low-temperature region, near the magnetic ordering in a static magnetic field of 0.42 T. The inverse magnetic susceptibility (the inset of Fig. 1) is a linear function of the temperature; in a temperature range of 100–290 K, it obeys the Curie–Weiss law. The paramagnetic Curie temperature θ_P of $TbNi_2$ compound is 37 K (the Curie constant is $C = 24.2$ K cm^3/g) and for the remaining samples θ_P decreases with increasing Sc content down to 31 K for $x = 0.2$. The values of the effective magnetic moment μ_{eff} calculated per terbium atom (the calculation were performed assuming that Ni atoms do not carry any magnetic moment) for all samples are very close to the value that is expected for a free Tb^{3+} ion and are equal to 9.6 μ_B. The Curie temperatures decrease from 36 K for $TbNi_2$ to 31 K for $Tb_{0.8}Sc_{0.2}Ni_2$.

* corresponding author; e-mail: cwikjac@ml.pan.wroc.pl

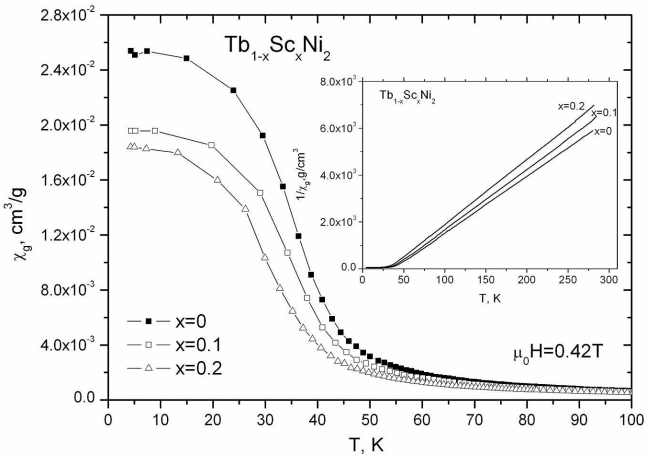

Fig. 1. Low temperature region dependence of the magnetic susceptibility of $Tb_{1-x}Sc_xNi_2$ ($x = 0$, 0.1, 0.2) solid solutions measured in 0.42 T. The inset shows the inverse magnetic susceptibility vs. temperature.

As an example, the temperature dependence of the heat capacity $C_{\rm tot}(T)$, the sum of electron and phonon $C_{\rm el+ph}(T)$ and magnetic contributions of solid solution $Tb_{0.9}Sc_{0.1}Ni_2$ are shown in Fig. 2. Shown is the tem-

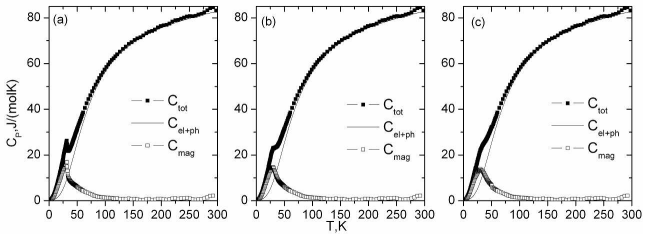

Fig. 2. The heat capacity of $Tb_{0.9}Sc_{0.1}Ni_2$ measured in zero (a), 0.42 T (b) and 1 T (c) magnetic field.

perature dependence of the heat capacity measured in zero magnetic field (Fig. 2a), 0.42 T (Fig. 2b) and 1 T (Fig. 2c). The sharp λ-like maximum on the heat capacity curves at $T_{\rm C} = 31$ K corresponds to the magnetic ordering temperature. In order to estimate the magnetic contribution a theoretical calculation of the Debye function was made [7]. The best fit for the wide temperature range of 2–300 K could be obtained by fixing the parameters $\gamma = 38$ mJ/(mol K^2) for all the measured samples, while the Debye temperature increases with increasing scandium content from $\theta_{\rm D} = 262$ K ($TbNi_2$) to $\theta_{\rm D} = 289$ K ($Tb_{0.8}Sc_{0.2}Ni_2$). Heat capacity measurements in magnetic fields show that the magnetic field causes the broadening and displacement of the λ-kind maximum. In order to calculate the MCE the magnetic part of the entropy was calculated by integrating $C_{\rm mag}(T)/T$. The magnetocaloric effect was calculated by a method suggested by von Ranke et al. [8]. We determined the temperature dependence of the adiabatic temperature change $\Delta T_{\rm ad}$ for measured $Tb_{1-x}Sc_xNi_2$ ($x = 0.1$, 0.2) in the maximum magnetic field of 1 T. For $\mu_0 H = 1$ T, the maximum magnetocaloric effect $\Delta T_{\rm ad}$ reaches about 2.4 K near 36 K for $TbNi_2$. In the case of the solid solutions the maximum $\Delta T_{\rm ad}$ reaches about 2 K near 33 K for $Tb_{0.9}Sc_{0.1}Ni_2$.

4. Conclusion

The effect of partial replacement of Tb by Sc on the structure and physical properties of the $Tb_{1-x}Sc_xNi_2$ solid solutions has been studied. The substitution results in the decrease in the ordering temperature, as observed both by the magnetization and specific heat measurements. The Curie temperature decreases from 36 K for $TbNi_2$ to 31 K for $Tb_{0.8}Sc_{0.2}Ni_2$. The moderate magnetocaloric effect in a magnetic field of 0.42 and 1 T is not very attractive for magnetic cooling.

References

[1] K.H.J. Buschow, in: *Ferromagnetic Materials*, Ed. E.P. Wohlfarth, Vol. 1, North Holland, Amsterdam 1980.

[2] W.E. Wallace, E. Segal, in: *Rare Earth Intermetallics*, Eds. A.M. Alper, J.L. Margrave, A.S. Nowick, Academic Press, New York 1973, p. 111.

[3] H.R. Kirchmayer, E. Burzo, in: *Landolt-Börnstein New Series III/19d2*, Ed. H.P.J. Wijn, Springer, Berlin 1990.

[4] E. Gratz, E. Goremychkin, M. Latroche, G. Hilscher, M. Rotter, H. Müller, A. Lindbaum, H. Michor, V. Paul-Boncour, T. Fernandez-Diaz, *J. Phys., Condens. Matter* **11**, 7893 (1999).

[5] J. Ćwik, Ph.D. Thesis, International Laboratory of High Magnetic Fields and Low Temperatures of PAS, Wrocław 2006.

[6] J. Ćwik, T. Palewski, K. Nenkov, J. Lyubina, J. Warchulska, J. Klamut, O. Gutfleisch, *J. Magn. Magn. Mater.* **321**, 2821 (2009).

[7] M. Bouvier, P. Lethuillier, D. Schmitt, *Phys. Rev. B* **43**, 13137 (1991).

[8] P.J. von Ranke, D.F. Grangera, A. Caladas, N.A. de Olivera, *J. Appl. Phys.* **93**, 4055 (2003).