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Magneto-History Effect in the $Tb_xGd_{1-x}Ni_3$ Compounds

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The compounds $Tb_xGd_{1-x}Ni_3$ with a $PuNi_3$-type structure have been obtained. The magnetic properties have been investigated by using SQUID magnetometer (Quantum Design MPMS, temperature from 1.9 K to 300 K and magnetic field up to 7 T). The partial replacement of Gd by Tb atoms is reflected in a decrease of the ordering temperature from 115 K ($x = 0.0$) to 81 K ($x = 1.0$) as well as the increase of the saturation magnetic moment M_S from 6.93 μ_B/f.u. ($x = 0.0$) to 7.14 μ_B/f.u. ($x = 1.0$). A large difference of $M(T)$ curves has been noticed between the so-called field cooling–zero field cooling magnetization. The thermomagnetic curves are sensitive to the applied magnetic field and their origin can be understood as the domain-wall pinning effect and as the temperature dependence of coercivity.

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1. Introduction

The RNi_3 compounds where R is rare earth show interesting properties due to the combination of $3d$ itinerant and $4f$ localized magnetism [1–7] and are quite useful materials for hydrogen storage [8]. Partially replacing of Ni atoms by other $3d$ elements or Gd by other $4f$ elements leads to some changes in magnetic properties correlated with their electronic structure [1–4, 7–10]. Previously studied $Tb_xGd_{1-x}Ni_3$ series crystallize in the $PuNi_3$-type of crystal structure and indicates the decrease of the Curie temperature from 115 K ($x = 0.0$) to 79 K ($x = 1.0$) [5]. Moreover for Tb-doped compounds there is observed second magnetic phase transition in lower temperature range T_h. As it was reported by Hashimoto *et al.* [4] the additional phase transition for $TbNi_3$ compound is usually explained by non-collinear magnetic structure of the Tb atoms confirmed by the neutron diffraction studies. The magnetic moments of terbium atoms in the same crystallographic position indicate ferromagnetic arrangement but they are turned between two different positions [4]. Thus, the magnetic transition observed at T_h can be connected with the canted terbium structure. For all studied $Tb_xGd_{1-x}Ni_3$ compounds the magnetic susceptibility follows the modified Curie–Weiss law and the values of effective magnetic moment (μ_{eff}) obtained from the Curie constant are higher than for free Gd^{3+} and Tb^{3+} ions. The excessive μ_{eff} can be the result of polarization Ni $3d$ by R $5d$ band. The values of the saturation magnetic moment (M_S) increase from 6.93 μ_B/f.u. ($x = 0.0$) to 7.14 μ_B/f.u. ($x = 1.0$). The change of magnetic entropy $\Delta S_m(T, H)$ exhibits oscillating behaviour. For Tb-rich compounds the magnetocaloric effect (MCE) effect at T_h is higher than at T_p. This behaviour can be explained by an increase in a magnetic disorder in $4f$ magnetic sublattice and canted structure of terbium [5]. The interesting magnetic properties of $Tb_xGd_{1-x}Ni_3$ system were the motivation for their precise study. Therefore in this paper we are focused on the influence of terbium substitution on the magneto-history effect of the $Tb_xGd_{1-x}Ni_3$ series.

2. Experimental details

The polycrystalline samples $Tb_xGd_{1-x}Ni_3$ ($x = 0.0$, 0.5, 0.8, 1.0) were prepared by arc-melting method from high purity elements under argon atmosphere. The samples were remelted several times and afterwards were wrapped in tantalum foil, placed in quartz tubes and annealed at 900 °C for one week. The crystal structure of all samples was checked by the means of X-ray diffraction (XRD) using Siemens D5000 diffractometer. The magnetic properties of examined samples were measured with the use of SQUID magnetometer (MPMS XL7 Quantum Design). All measurements were performed in the 2–300 K temperature range and magnetic field up to 7 T.

3. Results and discussion

The temperature dependence of magnetization $M(T)$ has been measured at different applied magnetic fields (10 Oe $< H_{appl} <$ 1000 Oe). The Curie temperature (T_C) has been estimated from dM/dT and decreases when Tb content increased from 115 K ($x = 0.0$) to 82 K ($x = 1.0$). In the magnetically ordered state all doped compounds exhibit remarkable thermomagnetic irreversibility (Fig. 1a–c). In the field cooled (FC) mode the $M(T)$ dependence measured at $H_{appl} \leq 0.1$ T indicates typical ferromagnetic behaviour and rises in low

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temperature range up to saturation. However, in the zero field cooled (ZFC) mode the $M(T)$ curve is quite different and the magnetization indicates smaller values at lower temperatures. Around the Curie temperature both curves overlap each other and this temperature is called as freezing or blocking temperature (T_F). The thermomagnetic FC–ZFC dependence can be considered as the magneto-history effect is mostly connected with the presence of narrow domain walls as well as the anisotropy of each ions. For the Gd ion the orbital momentum $L = 0$ and anisotropy can be neglected. However, for Tb ion $L = 3$ and the strong thermomagnetic dependence on $M(T)$ is observed, especially at $H_{\rm appl} = 10$ Oe (Fig. 1d). Additionally, for $x = 1.0$ a negative value of $M(T)$ is evidenced at low temperature range. Such behaviour can be considered as an influence of the anisotropy which may lead to a negative $M(T)$ value at quite relatively small applied magnetic field. Moreover, this effect may be also observed due to the applied demagnetization procedure in the vicinity of the magnetocrystalline anisotropy [11].

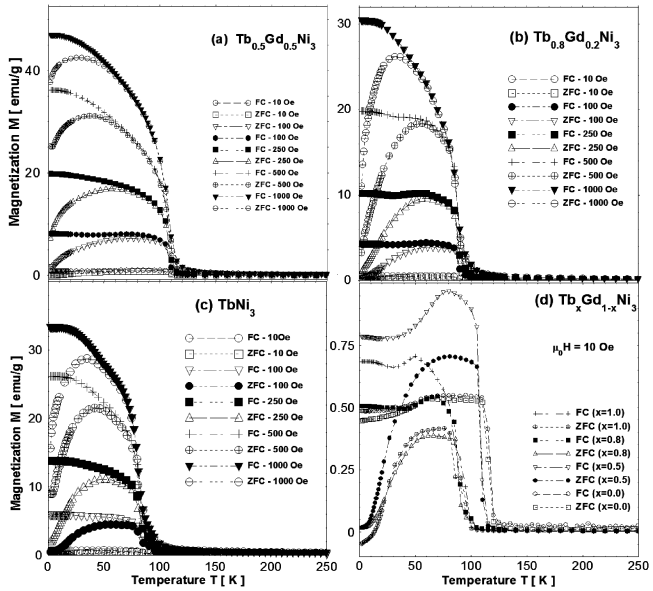

Fig. 1. The temperature dependence of FC–ZFC magnetization for the $Tb_xGd_{1-x}Ni_3$ system: (a) $x = 0.5$; (b) $x = 0.8$; (c) $x = 1.0$; (d) the comparison between FC–ZFC curves at $H_{\rm appl} = 10$ Oe.

A difference in FC–ZFC curves can be understood as the domain-wall pinning effect and as the temperature dependence of coercivity. During FC process the initial magnetic state of sample is obviously quite different than in ZFC process. In the zero applied magnetic field the magnetic domains are already arranged in random directions after cooling the sample below T_C. The value of magnetization at low temperatures remains small even though the small magnetic field is applied ($H_{\rm appl}$). It is closely connected with a quite high coercivity (H_C). In the FC process the magnetic domains are oriented along the $H_{\rm appl}$ direction when the sample is cooled. The domain walls move easier when the temperature increases and H_C is reduced. In a consequence an increase of the magnetization is observed. So, the magnetization in FC mode is larger than in ZFC.

The coercivity is the measure of the anisotropy according to the relation: $M_{\rm FC} - M_{\rm ZFC} \approx M_{\rm FC}H_C/(H_{\rm appl} + H_C)$ [12]. Thus, when the anisotropy increases with the Gd/Tb substitution then obviously the coercivity also increases. However, if the applied magnetic field ($H_{\rm appl}$) is higher and significantly exceeds the coercivity H_C, then the thermoremanent effect will disappear. In our case this effect is still observed at 0.1 T for all $x > 0.0$ samples whereas it is almost invisible in the $GdNi_3$ compound. So, the domain wall pinning is still observed at this field and higher $H_{\rm appl}$ is required to overcome such effect.

Moreover, the study of the hysteresis loops is a good and sensitive method of analysis of the domain motion effect and the influence of anisotropy. So, a series of measurements at different temperatures (2 K, 10 K, 25 K, 50 K, 100 K and 150 K) has been performed.

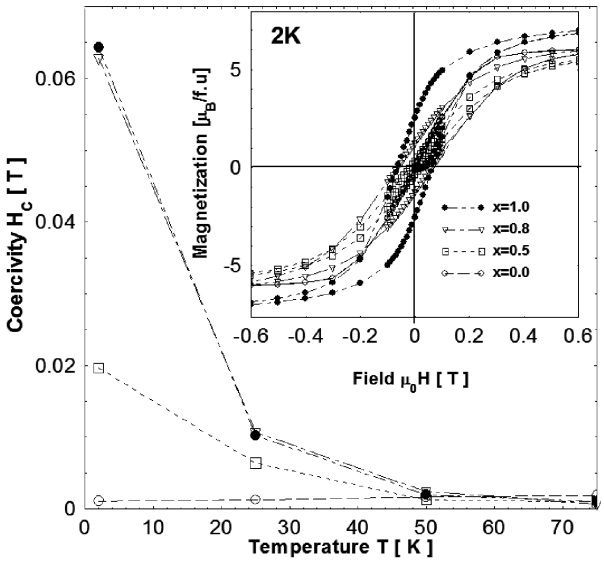

Fig. 2. The temperature dependence of coercivity H_C in the $Tb_xGd_{1-x}Ni_3$ system. Inset represents the hysteresis loops and initial magnetization curve at 2 K.

A significant hysteresis is observed at low temperatures. Therefore, the largest value of the coercive field H_C has been observed at 2 K (see Fig. 2) and decreases quickly with temperature. Moreover, a coercivity at low temperature is much more smaller in Gd-rich compounds as compared with larger amount of terbium. Hence, the observed magneto-history effect is more significant in the compounds with higher anisotropy and higher coercivity. It is well known that the propagation of narrow domain walls needs thermal activation. Additionally, the presence of narrow walls requires a large ratio of the anisotropy energy to the exchange energy. In our system the low T_C in $GdNi_3$ compound and the decrease of its values with increase of terbium content suggest that

the exchange interaction as well as the change energy are quite weak and become even weaker in doped compounds. Consequently, the ratio of the anisotropy energy to the exchange energy is higher in Tb-rich compounds and the magneto-history effect is stronger. The increase of the temperature leads to the decrease of coercivity. Since, the coercivity is the measure of the anisotropy then the anisotropy energy decrease rapidly with increasing temperature. As a consequence, the magneto-history effect vanishes.

Similar behaviour has been previously observed e.g in the RNi_2Mn [9, 10]. However, in our case the H_C in the whole studied system is almost ten times smaller than in the above system.

4. Summary

The series of the $Tb_xGd_{1-x}Ni_3$ intermetallic compound has been obtained. All synthesized compounds crystallize in the $PuNi_3$ rhombohedral structure. The Tb/Gd substitution is reflected in the decrease of the Curie temperature values from 115 K for $x = 0.0$ to 82 K for $x = 1.0$. The magneto-history effect has been evidenced for all Tb-doped compounds. The FC–ZFC curves indicate strong thermomagnetic dependence in the low applied magnetic field at low temperature range. The observed behaviour can be ascribed to the anisotropy as well as the temperature dependence of coercivity and indicates the presence of narrow domain walls formation.

Acknowledgments

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