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# Influence of Transition Metal Substitution on the Low-Field Magnetic Properties in the $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$ ( $\text{T} = \text{Fe}, \text{Co}$ ) Intermetallic Compounds

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The low-field magnetic properties of polycrystalline  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$  ( $\text{T} = \text{Fe}, \text{Co}$ ) intermetallic compounds are presented. The whole system crystallizes in the rhombohedral  $\text{PuNi}_3$  type of crystal structure. The composition dependence of the Curie temperature  $T_C(x)$  is associated with the change in the number of  $3d$  electrons. Moreover, the field cooled and zero field cooled (FC-ZFC) curves at low applied magnetic field are related to the anisotropy of  $\text{T}$  element. The saturation magnetic moment  $M_S(x)$  upon doping was estimated based on the hysteresis loops  $M(H)$ .

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## 1. Introduction

The  $RT_3$  compounds where  $R$  is rare earth and  $T$  is a transition metal have been previously studied very intensively [1–7]. The whole series indicate very interesting magnetic properties due to the combination of  $3d$  itinerant and  $4f$  localized magnetism. Among quite large group of the  $RT_3$  compounds one of the most known is  $R\text{Ni}_3$ . Partially replacing of Ni atoms by other  $3d$  elements leads to changes in magnetic properties correlated with the electronic structure [1–7]. One of previously studied series is  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$ , where  $\text{T} = \text{Fe}$  or  $\text{Co}$  [5, 8–11]. The doping in the  $3d$  sublattice the same amount of Co or Fe with different ionic radius (Fe — 0.77 Å, Co — 0.74 Å) and different number of  $3d$  electrons (Fe —  $3d^6$ , Co —  $3d^7$ ) causes appearing of different local environment of R atoms. It is reflected in the change of: the volume of the unit cell, the Curie temperature, the saturation magnetization, the magnetization on  $3d$  atom, the magnetocaloric effect (MCE) and the intensity of states at the Fermi level [8–11].

Here we present on influence of transition metal element substitution on the low-field magnetic properties of the  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$  ( $\text{T} = \text{Fe}, \text{Co}$ ) compounds.

## 2. Experimental details

The  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$ , where  $\text{T} = \text{Fe}, \text{Co}$  and  $x = 0.05, 0.1, 0.2, 0.6$  polycrystalline samples were prepared by arc-melting from high purity elements under argon atmosphere. The melted samples 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 means of X-ray diffraction (XRD) using Siemens D5000

diffractometer. The magnetic properties were carried out with the use of SQUID magnetometer (MPMS XL7 Quantum Design) in the range of 2 K–400 K. The AC magnetization was measured at zero applied magnetic field and at frequency 110 kHz. The DC magnetization was measured in FC and ZFC modes.

## 3. Results and discussion

The temperature dependence of AC and DC ( $\mu_0 H_{DC} = 10$  Oe) magnetization compounds is shown in Fig. 1. The Curie temperatures for  $x \leq 0.2$  are defined at the maximum of  $|dM/dT|$ . The values of  $T_C$  estimated at  $\mu_0 H_{DC} = 10$  Oe and  $\mu_0 H_{AC} = 0$  T modes are close to those previously obtained at 1000 Oe [8–11]. The change of  $T_C$  values with increasing the T content is larger in the case of Fe compounds. Thus,  $T_C$  in the iron series increases from 199 K ( $x = 0.05$ ) to 383 K to ( $x = 0.2$ ) respectively (see Table I). For the same cobalt concentration  $T_C$  varies more slowly. The  $T_C(x)$  dependence strongly depends on the electronic structure and filling of the  $3d$  band [9, 10] and can be explained similarly to other  $RT_3$  series on the basis of the rigid band model [1, 2, 8–11]. For higher  $x$  there is visible additional in the vicinity of  $T_C$ . The presence of such peak was evidenced in AC but also it was confirmed by zero field cooled (ZFC) measurements. The origin of this peak can be correlated with the Hopkinson effect. This effect is known since 1889 year and describes a phenomenon in which magnetic materials at low applied field reaches a maximum at a temperature  $T_H$  a just below the Curie temperature [13–15]. It was reported, that the presence of  $T_H$  is closely related to the initial susceptibility  $\chi_i$  which can be ascribed as  $\chi_i = M_S/\sqrt{K_1}$ , where  $K_1$  is the crystalline magnetic anisotropy constant and  $M_S$  denotes the saturation magnetization. In magnetic materials both parameters decrease with the increase of the temperature. However, close to  $T_C$  the anisotropy constant decreases

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faster than  $M_S$ . The change of  $K_1$  and  $M_S$  is reflected in the appearance of the Hopkinson peak when  $K_1 \rightarrow 0$ . Thus, such peak is often visible in systems with quite high FC-ZFC difference and large anisotropy. Similar behaviour was already observed e.g. in RPdIn [14], some soft magnetic materials such as Fe-Cu-Nb-Si-B [15] and Gd thin films [16].

TABLE I

Magnetic parameters for the  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$  ( $\text{T} = \text{Fe}, \text{Co}$ ) series. Data for: Fe — top, Co — down; 1 —  $T_C$  [K] from FC, 2 —  $T_C$  [K] from AC, 3 —  $T_H$  [K] from AC, 4 —  $M_S$  [ $\mu_B/\text{f.u.}$ ], 5 —  $M_{3d}$  [ $\mu_B/3d$  atom], 6 —  $M_{\text{FC-ZFC}}$  [emu/g], 7 —  $H_C$  [T], 8 —  $M_R$  [ $m\mu_B/\text{f.u.}$ ].

$x$	1	2	3	4	5	6	7	8
0.00	115	115	—	6.62	0.46	0.04	0.001	0.03
	115	—	—	6.62	0.46	0.04	0.001	0.03
0.05	199	198	—	6.38	0.21	0.02	0.002	0.09
	141	145	135	6.57	0.14	0.27	—	—
0.10	268	295	268	6.07	0.31	0.07	0.001	0.02
	165	170	160	6.21	0.26	0.73	0.003	0.04
0.20	381	383	—	5.46	0.51	0.17	0.001	0.03
	196	207	195	6.17	0.28	0.54	0.007	0.07
0.60	760 <sup>a</sup>	760 <sup>a</sup>	—	2.89	1.37	0.07	0.001	0.07
	339	355	340	5.03	0.66	0.36	0.008	0.02
1.00	725 <sup>a</sup>	725 <sup>a</sup>	—	1.61	1.8	0.05	0.001	0.02
	615 <sup>a</sup>	—	—	3.86	1.11	0.13	0.006	0.11

<sup>a</sup>data for:  $x = 0.60, 1.00$  (Fe) and  $x = 0.60$  (Fe) from Ref. [13, 16].

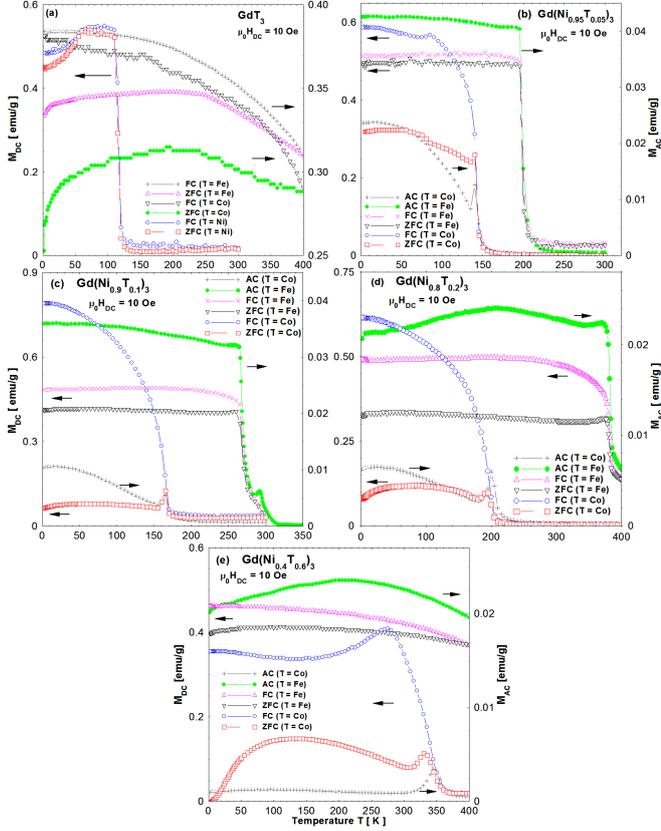


Fig. 1. The temperature dependence of DC magnetization for (a)  $\text{GdNi}_3$ ,  $\text{GdFe}_3$ ,  $\text{GdCo}_3$  as well as AC and DC magnetization for the  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$  ( $\text{T} = \text{Fe}, \text{Co}$ ) series (b)  $x = 0.05$ ; (c)  $x = 0.1$ ; (d)  $x = 0.2$ ; (e)  $x = 0.6$ .

The temperature dependence of magnetization at 10 Oe has been measured at two different modes. First in the field cooled mode (FC) and next in zero field cooled mode (ZFC). In the ordered state both  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$  systems exhibit a remarkable thermomagnetic irreversibility. Such behaviour is observed in many spin glasses but also in the ferromagnetic systems [17, 18]. The FC-ZFC dependence is mostly connected with the magnetic anisotropy and with the pinning domain effect. In the ZFC mode a pinning of domain walls is observed. The application of small magnetic field causes an alignment of some domains. When in such system the temperature is increased, then also an increase in magnetization is observed. In the vicinity of  $T_C$  a competitive process of magnetic moment reduction plays an important role. Thus a maximum in  $M(T)$  is visible. In FC mode the domains are saturated before pinning effect of domains

wall appears. So, in the  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$  studied series at low temperatures the  $M(T)$  in ZFC mode is smaller than in the FC mode. Moreover, the FC-ZFC difference ( $M_{\text{FC-ZFC}}$ ) is larger for  $\text{T} = \text{Co}$  than for  $\text{T} = \text{Fe}$ . Such behaviour is probably connected with the anisotropy of substituted element which is higher for Co content. However, the observed maximum in the  $M_{\text{FC-ZFC}}(x)$  dependence (Fig. 2) is probably connected with the behaviour of domain walls during cooling the samples with or without magnetic field.

The values of saturation magnetization ( $M_S$ ) were determined from the hysteresis loops (not shown here). Analyzing the  $M_S(x)$  and  $M_{\text{FC-ZFC}}(x)$  dependencies one can notice that both parameters are higher in the case of cobalt system. However, the second one indicates the maximum at low T element concentration (Fig. 2), which may be related to some changes in the magnetic interactions between transition metal elements having different anisotropy. The latter Ni/T substitution leads to the decrease of  $M_{\text{FC-ZFC}}$  value which is accompanied by the decrease of  $M_S(x)$ . The FC-ZFC irreversibility in  $M_{\text{DC}}(T)$  dependence has been previously described as:  $M_{\text{FC-ZFC}} \approx M_{\text{FC}}H_C/(H_{\text{appl}} + H_C)$ , where  $H_{\text{appl}}$  denotes the external applied magnetic field and  $H_C$  denotes coercivity. According to above formula, the observed FC-ZFC effect should disappear when  $H_{\text{appl}} \gg H_C$ .

For the  $\text{Gd}(\text{Ni}_{1-x}\text{T}_x)_3$  system the coercivity were determined directly from the hysteresis loops measured at 2 K. Its values are significantly low, but for higher Co content the  $H_C$  value is slightly higher than for Fe. Thus, for Co compounds we have observed stronger FC-ZFC effect. However, like in the RPdIn systems ( $\text{R} = \text{Tb-Er}$ ) in low temperature range antiferromagnetic interactions are dominant and anisotropy field is more important than coercivity field [14].

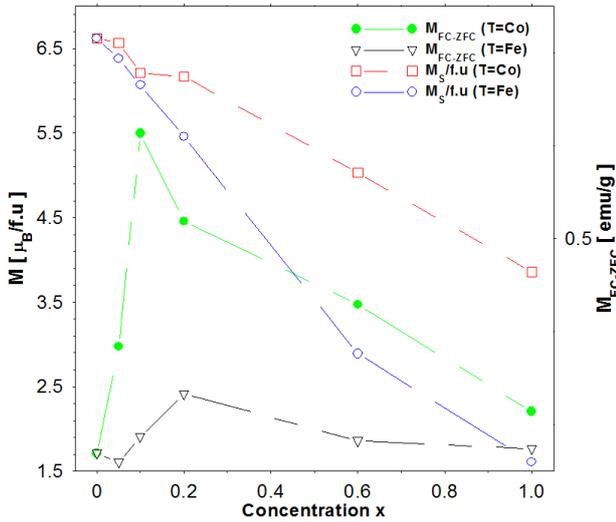


Fig. 2. The comparison between  $M_S(x)$  and  $M_{FC-ZFC}(x)$  dependencies in the  $Gd(Ni_{1-x}T_x)_3$  ( $T = Fe, Co$ ) series.

In our latter studies we have shown quite strong influence of the electronic structure of the substituted elements Ni/T where  $T = Co, Fe$  on the magnetic properties in the investigated compounds [1, 8, 11]. In the case of  $T = Fe$  the exchange splitting of 3d band was visible for Ni and Fe-rich concentration [1, 8]. For smaller iron doping the valence band exhibits the presence of only one broad peak. In the case of  $T = Co$  the reduction of 3d electrons with the increasing cobalt content causes the decrease of intensity of states near by the Fermi level but the valence band do not exhibit clearly the exchange splitting [8, 11]. Moreover, we did not observe maximum in the  $T_C(x)$  dependence like in the iron system. The rigid band model [2] which is suitable for explaining this behaviour is based on the density of states near by the Fermi level and filling of 3d bands. It is obvious that different number of 3d electrons in both studied systems is responsible for this behaviour.

Thus, Fe has one 3d electron less than Co and we have observed: i) maximum in  $T_C(x)$ , ii) lower  $M_S(x)$  but higher  $M_S/3d$  atom values, iii) lower  $M_{FC-ZFC}$  values. The opposite behaviour has been observed in the Co system. So, the different number of 3d electrons has an influence on magnetic  $fd$  interactions but also on the scattering mechanism which seems to be stronger in the case of iron than in cobalt compounds.

#### 4. Conclusions

The aim of presented work was to study low-field magnetic properties for  $Gd(Ni_{1-x}T_x)_3$  ( $T = Fe, Co$ ) series. It was evidenced that, the Ni/T substitution causes an increase of the Curie temperature  $T_C$ . However, the  $T_C$  increases faster in the case of Fe content. The increasing of  $x$  content leads to the appearing of Hopkinson peak in the AC and DC magnetization. The presence of

this peak is related to the saturation magnetization and anisotropy of studied systems. The anisotropy is higher for Co element, thus this behavior is well visible in the  $Gd(Ni_{1-x}Co_x)_3$  compounds. The FC-ZFC curves measured at 10 Oe revealed a quite strong thermomagnetic dependence. The  $M_{FC-ZFC}$  difference is larger in Co system than in Fe one. This kind of behaviour is related to the anisotropy of T elements.

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