Influence of Transition Metal Substitution on the Low-Field Magnetic Properties in the Gd(Ni1−xTx)x3 (T = Fe, Co) Intermetallic Compounds

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The low-field magnetic properties of polycrystalline Gd(Ni1−xTx)x3 (T = Fe, Co) intermetallic compounds are presented. The whole system crystallizes in the rhombohedral PuNi3 type of crystal structure. The composition dependence of the Curie temperature TC(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 T element. The saturation magnetic moment MS(x) upon doping was estimated based on the hysteresis loops M(H).

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1. Introduction

The RT3 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 RT3 compounds one of the most known is RNi3. 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 Gd(Ni1−xTx)x3, where T = Fe or 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 — 3d6, Co — 3d7) cause the appearance 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 Gd(Ni1−xTx)x3 (T = Fe, Co) compounds.

2. Experimental details

The Gd(Ni1−xTx)x3, where T = Fe, 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 (µ0HDC = 10 Oe) magnetization compounds is shown in Fig. 1. The Curie temperatures for x ≤ 0.2 are defined at the maximum of |dM/dT|. The values of TC estimated at µ0HDC = 10 Oe and µ0HAC = 0 Oe are close to those previously obtained at 1000 Oe [8–11]. The change of TC values with increasing the T content is larger in the case of Fe compounds. Thus, TC 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 TC varies more slowly. The TC(x) dependence strongly depends on the electronic structure and filling of the 3d band [9, 10] and can be explained similarly to other RT3 series on the basis of the rigid band model [1, 2, 8–11]. For higher x there is visible additional in the vicinity of TC. 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 TH a just below the Curie temperature [13–15]. It was reported, that the presence of TH is closely related to the initial susceptibility χ, which can be ascribed as χ = M2/K,T, where K1 is the crystalline magnetic anisotropy constant and M2 denotes the saturation magnetization. In magnetic materials both parameters decrease with the increase of the temperature. However, close to TC 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 $K_1 \to 0$. 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].

![Image](image-url)

**Fig. 1.** The temperature dependence of DC magnetization for (a) GdNi$_3$, GdFe$_3$, GdCo$_3$ as well as AC and DC magnetization for the Gd(Ni$_{1-x}$T$_x$)$_3$ (T = Fe, 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 Gd(Ni$_{1-x}$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 Gd(Ni$_{1-x}$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 ($M_{FC-ZFC}$) is larger for T = Co than for T = Fe. Such behaviour is probably connected with the anisotropy of substituted element which is higher for Co content. However, the observed maximum in the $M_{FC-ZFC}(x)$ 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 $M_{FC-ZFC}(x)$ 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 $M_{FC-ZFC}$ value which is accompanied by the decrease of $M_S(x)$. The FC-ZFC irreversibility in $M_{FC}(T)$ dependence has been previously described as:

$M_{FC-ZFC} \approx M_{FC}(HC)/(H_{app}+HC)$, where $H_{app}$ denotes the external applied magnetic field and $HC$ denotes coercivity. According to above formula, the observed FC-ZFC effect should disappear when $H_{app} \approx HC$.

For the Gd(Ni$_{1-x}$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 $HC$ value is slightly higher than for Fe. Thus, for Co compounds we have observed stronger FC-ZFC effect. However, like in the RPdIn systems (R = Th-Er) in low temperature range antiferromagnetic interactions are dominant and anisotropy field is more important than coercivity field [14].

### Magnetic parameters for the Gd(Ni$_{1-x}$T$_x$)$_3$ (T = Fe, Co) series.

<table>
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<th>$x$</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
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<td>115</td>
<td>6.62</td>
<td>0.46</td>
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<td>0.001</td>
<td>0.03</td>
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<td>198</td>
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<td>0.002</td>
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<tr>
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<td>269</td>
<td>6.07</td>
<td>0.31</td>
<td>0.07</td>
<td>0.001</td>
<td>0.02</td>
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<tr>
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<td>383</td>
<td>5.46</td>
<td>0.51</td>
<td>0.17</td>
<td>0.001</td>
<td>0.03</td>
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<tr>
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<td>760$^b$</td>
<td>5.03</td>
<td>0.66</td>
<td>0.36</td>
<td>0.008</td>
<td>0.02</td>
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</tr>
<tr>
<td>1.00</td>
<td>725$^a$</td>
<td>725$^b$</td>
<td>5.03</td>
<td>0.66</td>
<td>0.36</td>
<td>0.008</td>
<td>0.02</td>
<td></td>
</tr>
</tbody>
</table>

*data for: $x = 0.60, 1.00$ (Fe) and $x = 0.60$ (Fe) from Ref. [13, 16]
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.

References