Unusual magnetic properties of Yb\textsubscript{x}Gd\textsubscript{1-x}Ni\textsubscript{5} (x≤0.5) intermetallics

A. Bajorek\textsuperscript{1}, G. Chełkowska, A. Chrobak

A. Chełkowski Institute of Physics, University of Silesia, Uniwersytecka 4, 40-007 Katowice, Poland

E-mail: anna.bajorek@us.edu.pl

Abstract. In the presented paper we are focused on the magnetic properties of the hexagonal Yb\textsubscript{x}Gd\textsubscript{1-x}Ni\textsubscript{5} intermetallic compounds. Based on wide-ranging SQUID magnetometer (Quantum Design MPMS, temperature from 1.9K to 400K and magnetic field up to 7T) series of different magnetic measurements were carried out. It was shown that saturation magnetization and the Curie temperature strongly depends of Yb concentration. Moreover, the so-called field cooling - zero field cooling (FC-ZFC) curves reveal an unexpected existence of negative magnetization $M$ in a certain temperature range. The possible origin of the diamagnetic behaviour, observed in doped samples, has been rather ascribed to the interaction between more than one opposite aligned magnetic sublattices.

1. Introduction

The RNi\textsubscript{5} compounds (R - rare earth) have been intensively studied in recent decades [1-12]. The highest value of the Curie temperature ($T_c$) was evidenced for the GdNi\textsubscript{5} compound which exhibits magnetic ordering below about $T_c=30K$ [1-5]. Quite interesting properties show the newly obtained series of the Yb\textsubscript{x}Gd\textsubscript{1-x}Ni\textsubscript{5} compounds x≤0.5 [5], which crystallize in the hexagonal CaCu\textsubscript{5} type of crystal structure. The Yb/Gd substitution causes the increase of c lattice parameter in the whole range of studied concentrations. This kind of change is connected with different ionic radii for gadolinium and ytterbium ions. The deviation from the linear change of a(x) and V (x) for $x > 0.4$ could be related to intermediate valence of Yb ions. However, the gradual decrease of the residual resistivity value observed upon Yb/Gd substitution suggests the presence of mostly Yb\textsuperscript{3+} ions, which have smaller ionic radius than Yb\textsuperscript{2+}. The XPS valence band nearby the Fermi level for the whole series are dominated by Ni3d states. The multiplet structure in the valence bands is typical for Yb\textsuperscript{3+} states. Moreover, the valence of Yb ions is confirmed by the spectrum of the Yb4d core level line which is rather characteristic of Yb\textsuperscript{3+} structure. The properties of the Yb\textsubscript{x}Gd\textsubscript{1-x}Ni\textsubscript{5} compounds are very interesting and were the motivation to study their magnetic properties much more detailed.

2. Experimental

Polycrystalline samples of the Yb\textsubscript{x}Gd\textsubscript{1-x}Ni\textsubscript{5} series with x = 0.0, 0.2, 0.4, 0.5 were prepared by arc melting from high purity elements under argon atmosphere. The purity of materials used for preparing samples were 99.99% for Ni and Yb and 99.9% for Gd. The crystal structure of all samples was...
checked by means of X-ray diffraction (XRD) using Siemens D5000 diffractometer. The analysis of XRD patterns obtained at room temperature confirms that all as-cast studied compounds were single phase and crystallize in the hexagonal CaCu$_5$ structure [5]. The magnetic properties of Yb$_x$Gd$_{1-x}$Ni$_5$ compounds in a form of bulk regular pieces were measured with the use of SQUID magnetometer (MPMS XL7 Quantum Design). All measurements were performed in the 2K-400K temperature range up to 7T magnetic field. In the case of low field magnetic measurements a special attention was paid for discharging of the superconducting magnet by applying a demagnetizing procedure. In this procedure the magnetic field was alternated from 30 000 Oe to 1 Oe and the remanence was controlled by the means of a soft ferromagnet with low coercive field (about 2 A/m). After applying the demagnetizing procedure the magnet remanence was reduced from about 18 Oe to 1.5 Oe and was taken into account during setting the magnetic field.

### 3. Results and discussion

For the characterization of the magnetic ordering of the studied compounds the DC magnetization measurements have been carried out below the room temperature. Figure 1 presents the temperature dependence of DC magnetization $M(T)$ for all investigated samples measured at 0.1T of applied magnetic field. The Curie temperatures $T_C$ are defined at the maximum of $|dM/dT|$. The $T_C$ values for the whole series decrease significantly with increasing of Yb content. Indeed for $x=0.0$, $0.2$, $0.4$, $0.5$ the corresponding $T_C$ were found to be 33K, 28K, 20K, 16K respectively. Moreover, these results are consistent with the previously obtained from AC measurements making with the use of AC magnetometer as also from the electrical resistivity investigations [5]. The $M(T)$ curves at this 0.1 T applied field exhibit typical behaviour, raises rapidly with lowering the temperature and saturates in low temperatures. The $T_C$ values were also estimated from the Arrott plots. Figure 2 represents these plots for $x=0.4$ ytterbium concentration. One can notice that the value of $T_C$ is close to that obtained from $M(T)$ curve. Similar results were received for other studied compounds. The magnetization curves for $M(H)$ the Yb$_x$Gd$_{1-x}$Ni$_5$ system measured at 2K and up to 7T saturate faster for the Gd – rich compounds. The highest value of the saturation magnetization $M_S$ is observed for $x=0.0$ (6.52 $\mu_B$/f.u) and slowly decreases with the increasing of ytterbium concentration up to $x=0.5$ (4.36 $\mu_B$/f.u). The value of $M_S$ for the GdNi$_5$ compound is smaller than for free Gd$^{3+}$ ion (7 $\mu_B$). This kind of behaviour is connected with a negative polarization of the Ni3d band by Gd atoms, which leads to reduction of the magnetic moment.

![Figure 1. The temperature dependence of DC magnetization M(T) in the Yb$_x$Gd$_{1-x}$Ni$_5$ series measured at 0.1T magnetic field.](image1)

![Figure 2. The Arrott plots for the Yb$_{0.4}$Gd$_{0.6}$Ni$_5$ compound.](image2)
Quite interesting is the temperature dependence of magnetization $M(T)$ measured at different magnetic fields with the use of the so-called zero field cooled (ZFC) – field cooled (FC) procedure. One can notice that $M(T)$ behaviour for the GdNi$_5$ is different than that for doped compounds. In the ordered state samples with $0.2 \leq x \leq 0.5$ exhibit a remarkable thermomagnetic irreversibility that is depicted in Figure 3. This feature is better demonstrated for lower magnetic field (below 500 Oe). For the samples with $x \geq 0.2$ above $T_C$ the magnetization has a minimum at certain temperature and (in some cases) exhibits a negative value. Moreover, this negative magnetization depends on the magnetic field and on the way of measurements (FC or ZFC). With the increasing of applied magnetic field the position of the minimum $M(T)$ moves into higher temperatures and its value changes from negative to positive. The $M(T)$ dependence also changes with the ytterbium concentration.

The unexpected behavior i.e. the appearing of negative magnetization is the strongest for $x=0.4$ ytterbium concentration. The zero field cooled (ZFC) sample behaves normally without any trace of negative $M(T)$ value in low temperature range. The temperature dependence of magnetization $M(T)$ for the field cooled (FC) sample shows quite different character. The negative magnetization is observed at about 20 – 70 K temperature range in 10 Oe and 100 Oe magnetic field. The minimum is followed by a broad maximum in temperature about 100 K. The value of this maximum increases with increasing an applied magnetic field. It should be stressed that similar results were obtained for the sample with $x=0.5$ that can be a confirmation of the presented effect and its dependence on ytterbium concentration. The observed negative values of $M(T)$ in the low magnetic fields at FC mode are

\[ M(T) \]

Figure 2. The temperature dependence of magnetization $M(T)$ for the Yb$_x$Gd$_{1-x}$Ni$_5$ compounds where: (a)$x=0.0$, (b) $x=0.2$, (c) $x=0.4$ and (d)$x=0.5$. 
difficult for interpretation. Such effect was already observed for several intermetallic compounds in a low magnetic field and low temperature range under the FC conditions [13,14]. The authors suggest that this kind of behaviour can be the result of inhomogeneous distribution of the magnetic moment in the samples or a substantial crystallographic disorder. In our case the unusual magnetic behaviour is visible above $T_C$ which is quite unexpected. We cannot explicitly claim that $T_C$ is only one temperature of magnetic phase transition. Thus, the performed measurements do not give a clear explanation of revealed behaviour so more much precise magnetic, structural and transport measurements are required.

4. Conclusions

The main conclusions of the presented work can be summarized as follow:

- Partial substitution of Yb for Gd atoms in the Yb$_x$Gd$_{1-x}$Ni$_5$ series ($x \leq 0.5$) causes the decrease of the Curie temperature from 33K to 16K and the decrease of the saturation magnetization from $6.52 \mu_B$/f.u to $4.36 \mu_B$/f.u for $x=0$ to $x=0.5$ respectively.

- With the increase of Yb concentration in the Yb$_x$Gd$_{1-x}$Ni$_5$ series an unusual $M(T)$ dependence i.e. the presence of negative magnetization is observed. The $M(T)$ values are very sensitive to applied magnetic field and the way of measurements (ZFC, FC). The consequence of Yb/Gd substitution could be the crystallographic disorder in the unit cell which can lead to a formation of more than one magnetic sublattices. It seems that in the favourable measurements conditions the interactions between these sublattices are reflected in the negative values of $M(T)$ dependence.

References

[1] Buschow K H J 1977 Rep. Prog. Phys. 40 1169
[2] Burzo E, Chełkowski A and Kirchmayr H R 1990 Landolt Börnstein Handbook, Springer Berlin vol.III/192
[3] Coldea M, Chiuzaian S G, Neumann M, Todoran D, Demeter M, Tetean R and Pop V 2000 Acta Physica Polonica A 98 629
[4] Gignoux D, Givord D and Del. Moral A 1976 Sol. State. Comm. 19 891
[5] Bajorek A and Chelkowska G 2009 Acta Physica Polonica A 115 188
[6] Burzo E, Pop V and Costina I 1996 J. Magn. Magn. Matter. 157/158 615
[7] Nesbitt E A, Williams H J, Wernick H J, and Sherwood R C 1962 J. Appl. Phys. 33 1674
[8] Bajorek A, Styśiak D, Chelkowska G, Deniszczyk J, Borgiel W and Neumann M 2006 Materials Science - Poland 24 867
[9] Balkis Ameen K 2002 J. Alloys. Compd. 347 165
[10] Fremy M A and Gignoux D 1985 J.of Less-Comm. Metals 106 251
[11] Palenzona A and Cirafici S 1973 J.of Less-Comm. Metals 33 361
[12] He J, Tsujii N, Nakanishi M, Yoshimura K and Kosuge K 1996 J. Alloys. Compd. 240 261
[13] Suski W 2007 Materials Science – Poland 25 333
[14] Toliński T, Andrzejewski B, Kowalczyk A, Chelkowska G, Szlaferek A and Frąckowiak J 2006 J. Phys. Chem. Solids 67 751