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Magnetic properties and resonance studies of some gadolinium-transition-metal-boron compounds

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In honor of Martin Peter's 60th birthday.

Abstract. The magnetic properties of ferrimagnetic $(Gd_yY_{1-y})_2Fe_{14}B$, $Gd_2Fe_{14-x}M_xB$ (M = Si or Ni), $(Gd_yY_{1-x})Co_4B$ and $Gd(Co_xNi_{1-x})_4B$ compounds are studied by magnetic measurements and ferromagnetic resonance. The iron and cobalt mean moments are dependent on composition. The effective splitting factors for $(Gd_yY_{1-y})Co_4B$ and $Gd(Co_xNi_{1-x})_4B$ compounds may be described by using the Vangsness's relation with $g_{Gd} = 2.00$ and $g_{Co} = 2.16$. Finally, the magnetic behaviour of transition metals in these compounds is discussed.

1. Introduction

Professor Peter Martin is one of the first who started the studies of rare-earth (R) compounds by means of resonance methods, his works in the field being classical [1, 2]. It is a pleasure to contribute to the special issue of Helvetica Acta Physica in honour of his sixtieth birthday, with a subject closely related to the above mentioned works.

Over the past four years the rare-earth-transition metal-boron compounds retained the attention of scientists. The discovery of a new material based on Nd-Fe-B alloy which could be used to produce permanent magnets with energy product up to 45 MGOe triggered a cascade of investigations devoted both to the understanding of the physical properties of these alloys and to improving the technological procedures of permanent magnets fabrications [3]. The intrinsic magnetic properties of these magnets are associated with the $R_2Fe_{14}B$ ternary phase. The study of other systems as those based on RCo_4B are also of interest in obtained information on the magnetic behaviour of transition metals in various hosts [4].

In the following we present some results concerning the magnetic properties of $(Gd_yY_{1-y})Fe_{14}B$, $Gd_2Fe_{14-x}M_xB$ (M = Si or Ni), $(Gd_yY_{1-y})Co_4B$ and $Gd(Co_xNi_{1-x})_4B$ systems. In addition, some compounds were studied by resonance method in order to obtain additional information on the above systems.

The X-ray analysis shows that $(Gd_yY_{1-y})_2Fe_{14}B$ compounds crystallize in a tetragonal structure belonging to the space group $P4_2/mnm$ in all the composition range [5]. This structure is retained in $Gd_2Fe_{14-x}M_xB$ systems having $x \leq 2.0$ for $M = Si$ and $x \leq 4.0$ for $M = Ni$. The $(Gd_yY_{1-y})Co_4B$ compounds crystallize in a $CeCo_4B$-type structure [6] as well as $Gd(Co_xNi_{1-x})_4B$ system for $x \geq 0.25$. For a smaller cobalt content, two phases are observed.

In $R_2Fe_{14}B$ compounds iron occupies six non-equivalent sites and cobalt in RCo_4B two types of sites. For a given site, the magnetic moment of the transition metals depends on the number and type of the nearest neighbour atoms [3]. By magnetic measurements only their mean values are obtained.

2. Magnetic properties of $Gd_2Fe_{14}B$-based compounds

The composition dependences of the magnetizations in $(Gd_yY_{1-y})Fe_{14}B$, $Gd_2Fe_{14-x}Si_xB$ and $Gd_2Fe_{14-x}Ni_xB$ compounds are plotted in Fig. 1. The magnetizations of $(Gd_yY_{1-y})_2Fe_{14}B$ increase when substituting gadolinium by yttrium. The magnetizations of $Gd_2Fe_{14}B$-based compounds decrease when iron is partially substituted by Si or Ni. These data show that all the studied systems are ferrimagnetically ordered. In the following we consider a two-sublattices ferrimagnet. This approximation neglects the differences in the magnetic moments of transition metal atoms located in various lattice sites, taking into account only

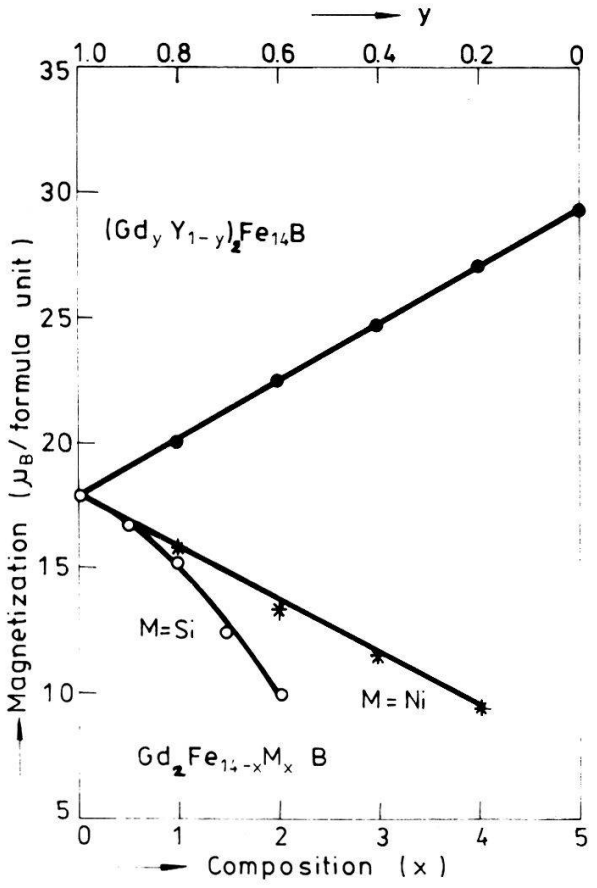

Figure 1
Composition dependence of the magnetization at 4.2 K in $(Gd_yY_{1-y})_2Fe_{14}B$ and $Gd_2Fe_{14-x}M_xB$ (M = Si or Ni) compounds

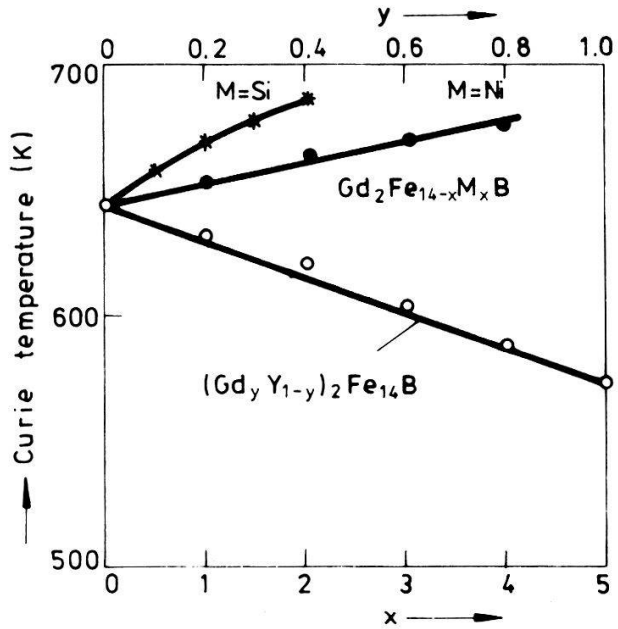

Figure 2
Composition dependence of the Curie temperature in $(Gd_yY_{1-y})_2Fe_{14}B$ and $Gd_2Fe_{14-x}M_xB$ (M = Si or Ni) compounds

their mean contributions. Admitting that the magnetic moment of gadolinium is $7\,\mu_B$, we determined the mean moments of the transition metals. In $(Gd_yY_{1-y})_2Fe_{14}B$ the iron's mean moment vary from $2.12\,\mu_B$ ($x = 0$) up to $2.26\,\mu_B$ ($x = 1.0$). In $Gd_2Fe_{14-x}Si_xB$, the iron's mean moment decreases down to $1.95\,\mu_B$ for $x = 2.0$, while in $Gd_2Fe_{14-x}Ni_xB$, the mean moment of the transition metal is $2.05\,\mu_B$, for $x = 4.0$

The composition dependences of the Curie temperatures, T_c, are plotted in Fig. 2. The T_c values increase when substituting Y by Gd. An increase in the Curie points is observed also when Fe is replaced by Si or Ni. When replacing iron by siilicon or nickel, generally a decrease in the exchange interactions exists and a lowered Curie point is expected. The data from Fig. 2 may be explained taking into account the crystal structure of $R_2Fe_{14}B$ compounds. In this lattice the distances between iron atoms cover a wide range of values. The interactions between iron atoms situated at distances $d \lesssim 2.45$ Å are negative while for larger distances are positive [7, 8]. Thus, the interactions between $Fe_3(8j_1)$ and $Fe_6(16k_2)$ atoms are strongly negative and those between $Fe_3(8j_1)$ and $Fe_3(8j_1)$ are also negative but having a smaller intensity. The magnetic couplings between $Fe_3(8j_1)$ and iron situated in $Fe_4(8j_2)$ and $Fe_5(16k_1)$ sites are positive and impose a parallel alignment of the iron moments [9]. The presence of the negative exchange interactions bring about the low ordering temperatures of $R_2Fe_{14}B$ compounds. The increase in the T_c values when replacing Fe by Si or Ni may be attributed only to the diminution of the negative exchange interactions. This takes place if Si or Ni atoms occupy Fe sites involved in antiferromagnetic exchange interactions, namely $Fe_6(16k_2)$ and $Fe_3(8j_1)$ sites.

3. Magnetic properties of $GdCo_4B$-based compounds

The composition dependences of the magnetizations at 4.2 K in $(Gd_yY_{1-y})Co_4B$ and $Gd(Co_xNi_{1-x})_4B$ compounds are plotted in Fig. 3. The

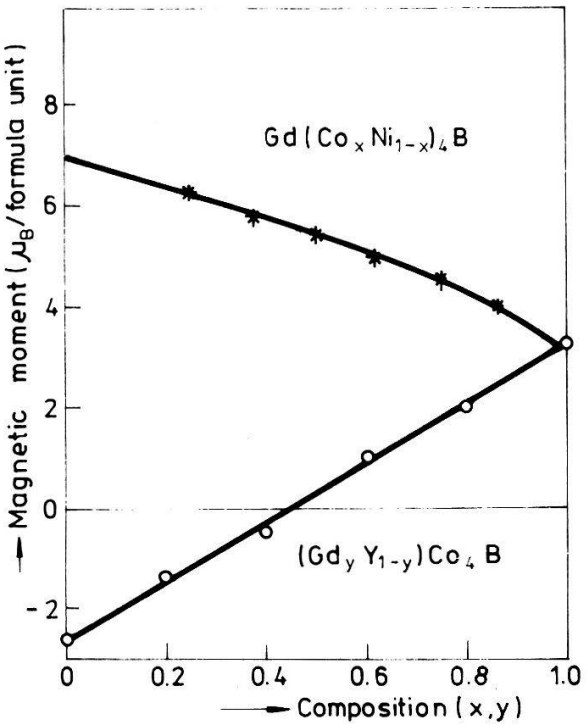

Figure 3
Composition dependence of the magnetization at 4.2 K in $(Gd_yY_{1-y})Co_4B$ and $Gd(Co_xNi_{1-x})_4B$ compounds

magnetizations of $(Gd_yY_{1-y})Co_4B$ system, decrease when gadolinium is substituted by yttrium from 3.25 μ_B ($x = 1.0$) up to a composition $x \simeq 0.43$, when the resultant magnetic moment is nil. Then the magnetizations increase up to 2.65 μ_B, a value determined in YCo_4B compound. In $Gd(Co_xNi_{1-x})_4B$ system, the magnetizations increase gradually when cobalt is substituted by nickel.

Admitting, as previously, that the magnetic moment of gadolinium is 7 μ_B we determined the mean cobalt moments, M_{Co}. In $(Gd_yY_{1-y})Co_4B$ compounds the M_{Co} values increase from 0.66 μ_B ($y = 0$) up to 0.94 μ_B ($y = 1.0$), as yttrium is substituted by gadolinium. In $Gd(Co_xNi_{1-x})_4B$ compounds the magnetic moment

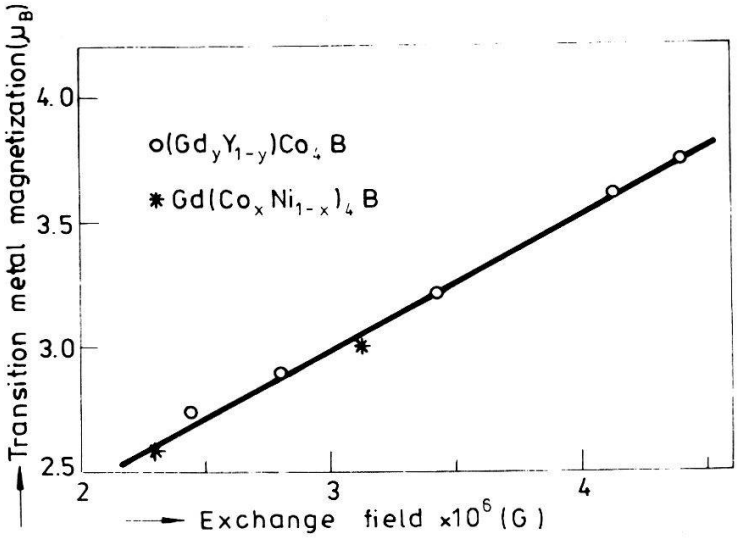

Figure 4
The dependence of the cobalt magnetization determined in $(Gd_yY_{1-y})Co_4B$ and $Gd(Co_XNi_{1-x})_4B$ compounds on the exchange fields

of the transition metals decreases up to $0.2\,\mu_B$ for $x = 0.25$. The extrapolation at $x = 0$ suggests a nearly zero magnetic contribution from the transition metals.

By using the magnetic measurements performed above the Curie points, we determined the exchange coefficients characterising the magnetic interactions inside and between the magnetic sublattices, as well as the exchange fields acting on the transition metal atoms. The dependence of the transition metal magnetizations on the exchange fields is plotted in Fig. 4. The data obtained for both $(Gd_yY_{1-y})Co_4B$ and $Gd(Co_xNi_{1-x})_4B$ systems are situated on the same curve. A linear relation of the form $M_{Co} \simeq bH_{ex}(Co)$ is obtained. The proportionality constant, b, is close to that determined in pseudobinary rare-earth-cobalt compounds [10]. This fact suggests that the variation of cobalt moment cannot be attributed to the band filling effects.

4. Ferromagnetic resonance studies

The composition dependences of the effective spectroscopic splitting factors g_{eff} for $(Gd_yY_{1-y})Co_4M$ and $Gd(Co_xNi_{1-x})_4B$ compounds are plotted in Fig. 5. The g_{eff} values are dependent on composition.

The resonance data evidenced in Fig. 5 may be analysed by using the Wangsness's relation [11]

$$g_{eff} = \frac{M_{Gd} - M_T}{M_{Gd}/g_{Gd} - M_T/g_T} \tag{1}$$

where M_{Gd} and M_T are the magnetizations of gadolinium and transition metal sublattices and g_{Gd} and g_T are their spectroscopic splitting factors, respectively.

Using the M_{Gd} and M_{Co} values obtained from magnetic measurements, we

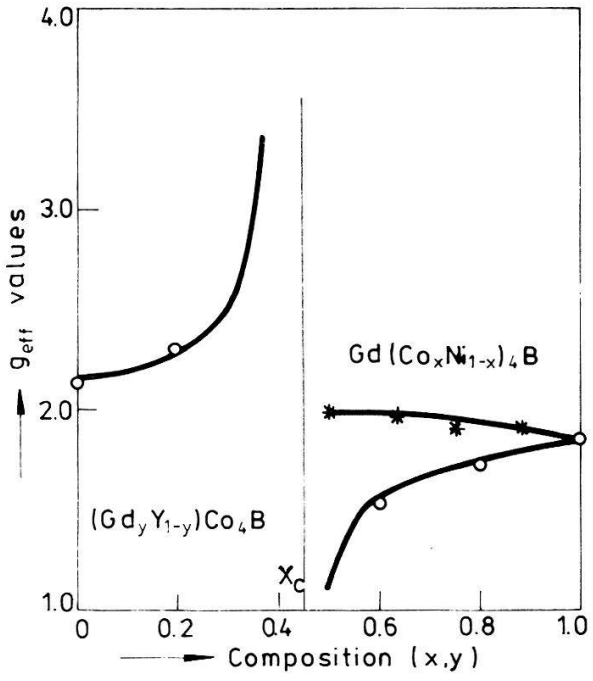

Figure 5
Composition dependence of the effective splitting factor in $(Gd_yY_{1-y})Co_4B$ and $Gd(Co_xNi_{1-x})_4B$ compounds

determined the expected g_{eff} values. We supposed that $g_{Gd} = 2.00$ and $g_{Co} = 2.16$. The calculated data, plotted in Fig. 5 by solid lines, are close to those experimentally determined. This shows that the Wangsness's relation describes well the ferromagnetic resonance behaviour of the studied compounds with g_{Co} value close to that determined in cobalt. Similar results have been obtained in $(Gd_yY_{1-y})Fe_2$ compounds [12].

The ferromagnetic resonance measurements on $Y_2Fe_{14}B$ compound show that the g value is $\simeq 2.08$, nearly the same as that evidenced in pure iron.

5. Discussion

The ferromagnetic resonance measurements show that in the studied compounds the g_{Co} and g_{Fe} values are close to the spectroscopic splitting factors of the pure metals. This fact suggests that the electronic configuration of the transition metals is not much altered by alloying. In addition to the linear dependence between cobalt moment and exchange field, the above behaviour suggests that the decrease in the transition metal moment is due to a reduced exchange splitting of their 3d states. Evidence of the above mechanism has been previously given [13] and was confirmed by the analysis of valence band spectra in Gd-Fe intermetallic compounds [14]. The iron moments are less sensitive to the exchange interactions than the cobalt ones, showing a more localized behaviour.

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