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LETTER TO THE EDITOR

## A comparison of the effect of pressure and Co substitution on the Néel temperature of the $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$ series

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**Abstract.** The effect of hydrostatic pressure and alloying on the magnetic ordering temperature in the  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  series was studied. The observed behaviour was related to changes in the density of states at the Fermi level caused by pressure or Ni substitution. In addition, the elastic constants of  $\text{YNi}_2\text{B}_2\text{C}$ , which is a representative member of the  $\text{RNi}_2\text{B}_2\text{C}$  family, were measured.

The recently discovered new family of quaternary borocarbides  $\text{RT}_2\text{B}_2\text{C}$  ( $\text{R} = \text{Y}$ , rare earth,  $\text{T} = \text{Ni}, \text{Co}, \text{Pd}, \dots$ ) [1–3] is attracting a lot of attention due to the elevated superconducting transition temperature for some of the members of the family ( $\text{R} = \text{Lu}, \text{Y}, \dots$ ) or the complicated magnetic phase diagram, and sometimes interplay between superconductivity and magnetism for the compounds with  $\text{R} =$  magnetic rare earth. Quaternary borocarbides crystallize in the  $\text{ThCr}_2\text{Si}_2$  type of crystalline structure with the alternation of  $\text{R-C}$  and (corrugated)  $\text{T-B}$  layers [4]. Magnetic interactions between  $\text{R-C}$  layers ( $\text{R} =$  magnetic rare earth) are considered to be of RKKY type mediated through transition metal d electrons [5]. Band structure calculations [6–8] show that for  $\text{RNi}_2\text{B}_2\text{C}$  the position of the Fermi level is close to the peak (formed mainly by Ni 3d electrons) in the density of states (DOS). It was argued that the DOS at the Fermi level or, in other words, the position of the Fermi level with respect to the peak in the DOS, can be a determinant factor for the changes of the superconducting transition temperature with the Ni-site substitution [9–11] or under pressure [12]. As far as we know, the effect of Ni-site substitution or pressure on magnetic properties of quaternary borocarbides has been studied less extensively: the Néel temperature ( $T_N$ ) was observed to increase under pressure in  $\text{HoNi}_2\text{B}_2\text{C}$  and  $\text{ErNi}_2\text{B}_2\text{C}$  [13–15] and decrease with Co substitution in  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  [16].

In the present work we combine Ni-site substitution with application of high pressure in the  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  series in an attempt to obtain information about different contributions to the changes in the Néel temperature in the series. Analysis of high-pressure results usually requires the knowledge of the elastic constants of the material under study. Room-temperature x-ray diffraction (XRD) measurements under pressure for one of the representatives of the quaternary borocarbide family ( $\text{YNi}_2\text{B}_2\text{C}$ ) were also performed as a part of our work.

Polycrystalline samples with the nominal composition  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$ ,  $x = 0, 0.25, 0.5, 0.75$  and 1, and  $\text{YNi}_2\text{B}_2\text{C}$  were prepared by a conventional arc-melting technique in partial argon atmosphere. Resistance measurements under pressure up to 16 kbar were

performed in the 2–300 K temperature range by a standard four-probe DC technique in a hydrostatic pressure cell similar to the one used in [17]. A 40:60 mixture of mineral oil and *n*-pentane was used as a pressure medium. The pressure at room temperature was measured *in situ* by a manganine manometer and the low-temperature values were obtained using the calibration against In or Sn superconducting manometers from [18]. Room-temperature XRD for the YNi<sub>2</sub>B<sub>2</sub>C sample under hydrostatic pressure up to 60 kbar was measured in a diamond anvil cell using the facilities of the Institute for High-Pressure Physics (see [19] for the experimental details).

Results for GdNi<sub>2</sub>B<sub>2</sub>C arc-melted polycrystalline samples [20] and single crystals [21], as well as for polycrystalline samples of the Gd(Ni<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>B<sub>2</sub>C series [16], suggest that more than one magnetically ordered state is present in the system under study. In this work we restrict ourselves only to the data related to the transition from the paramagnetic to the antiferromagnetic phase, which are well defined in both magnetic and transport properties.

XRD measurements at ambient pressure and temperature show that a ThCr<sub>2</sub>Si<sub>2</sub>-like crystalline structure was formed as a majority phase for all samples under study. The observed low ( $\leq 10\%$ ) level of impurities seems to be common for polycrystalline samples of the borocarbides prepared by arc melting. Small amounts of impurities in the samples as well as strains which necessarily exist in polycrystalline samples could, in principle, affect the results of the measurements under pressure. In the samples under study, the same procedure and starting materials were used for the whole series. The Néel temperature  $T_N$  obtained for GdNi<sub>2</sub>B<sub>2</sub>C is close to the one measured for the single crystal [21]. Both facts give us a hope that, at least semi-quantitatively, the evolution of the behaviour under pressure through the series is reflected correctly in the present results.

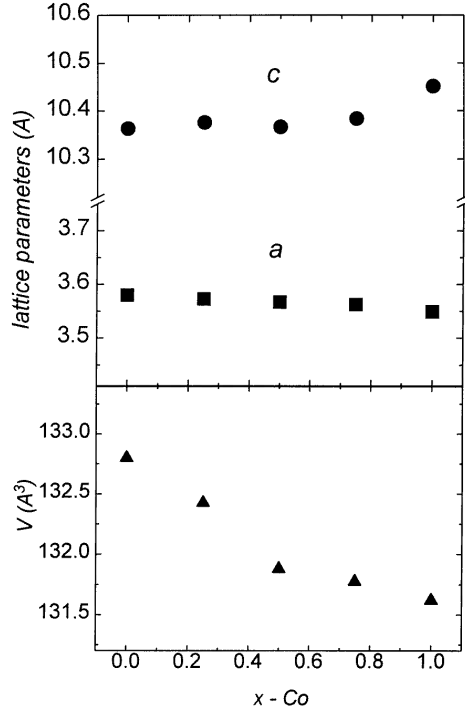
The variation of the lattice parameters and the unit cell volume with Co concentration in the Gd(Ni<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>B<sub>2</sub>C series is shown in figure 1. The changes in the lattice parameters and the unit cell volume are very small (i.e. the unit cell volume changes only by  $\Delta V/V_0 \approx -1\%$  between the end compounds GdNi<sub>2</sub>B<sub>2</sub>C and GdCo<sub>2</sub>B<sub>2</sub>C).

The lattice parameters and the unit cell volume under pressure for YNi<sub>2</sub>B<sub>2</sub>C, a representative member of the family of quaternary borocarbides, are shown in figure 2. The lattice parameters decrease linearly under pressure, giving the values of elastic constants  $d \ln a/dP = (-0.22 \pm 0.02) \times 10^{-3} \text{ kbar}^{-1}$ ,  $d \ln c/dP = (-0.40 \pm 0.04) \times 10^{-3} \text{ kbar}^{-1}$ ,  $d \ln V/dP = (-0.82 \pm 0.08) \times 10^{-3} \text{ kbar}^{-1}$ . These values are very close to the ones reported in [22] for CeRu<sub>2</sub>Si<sub>2</sub>—another compound with the ThCr<sub>2</sub>Si<sub>2</sub> type of crystalline structure. This suggests that rigid B–C bonds, present in borocarbides [23] (and absent from CeRu<sub>2</sub>Si<sub>2</sub>), do not affect drastically the changes of *c* lattice parameter under pressure. In the following we will assume that the elastic constants for the Gd(Ni<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>B<sub>2</sub>C series are the same as for YNi<sub>2</sub>B<sub>2</sub>C, as a reasonable approximation.

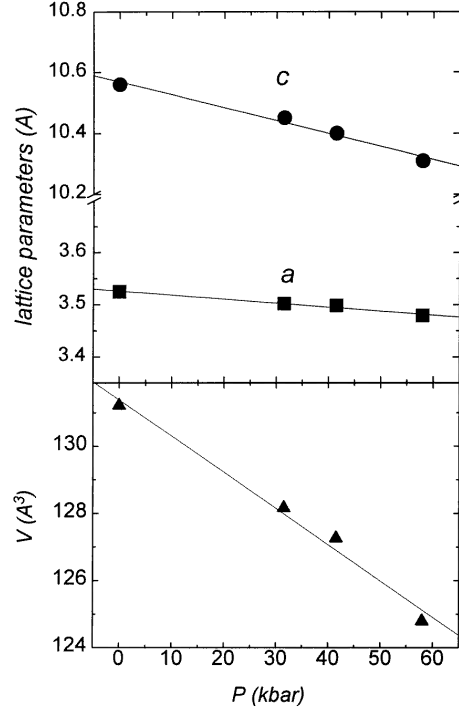
In figure 3, the low-temperature parts of the resistance curves, as a function of the temperature, are shown for Gd(Ni<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>B<sub>2</sub>C,  $x = 0, 0.5$  and  $1.0$ , at ambient pressure, from which the magnetic transition can be extracted.

The magnetic ordering temperature as a function of  $x$ , obtained from resistance measurements, is shown in figure 4. Substitution of Ni by Co causes a decrease of  $T_N$  with quite a sharp drop of the Néel temperature between  $x = 0.25$  and  $0.75$ . The composition dependences for both lattice parameters and  $T_N(x)$ , in the Gd(Ni<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>B<sub>2</sub>C series, are consistent with our previous data for another batch of samples [16].

The changes of the magnitude ordering temperature under pressure are small. The value of the pressure derivative  $dT_N/dP$  for pure GdNi<sub>2</sub>B<sub>2</sub>C is of the same order of magnitude as the ones measured in HoNi<sub>2</sub>B<sub>2</sub>C and ErNi<sub>2</sub>B<sub>2</sub>C [15]. However, an interesting feature of the pressure dependences in the Gd(Ni<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>B<sub>2</sub>C series is the change of sign of the



**Figure 1.** The lattice parameters and the unit cell volume as a function of Co concentration in the  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  series.



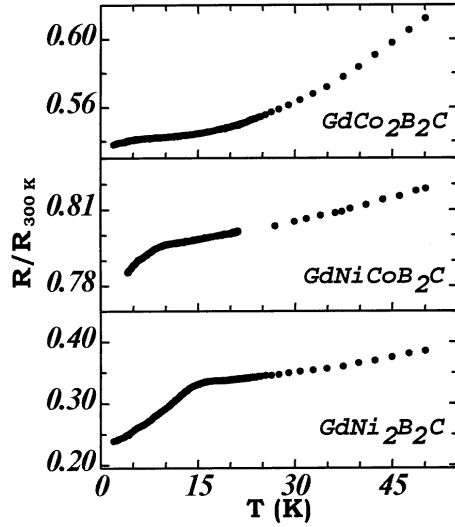
**Figure 2.** The pressure dependence of the lattice parameters and the unit cell volume for  $\text{YNi}_2\text{B}_2\text{C}$ .

pressure derivatives  $dT_N/dP$ , from positive for  $x \leq 0.5$ , to negative for  $x = 0.75$  and 1 (figure 5). So, the regions of Co concentration where the behaviour of  $T_N$  and its pressure derivative  $dT_N/dP$  display a fast variation practically coincide.

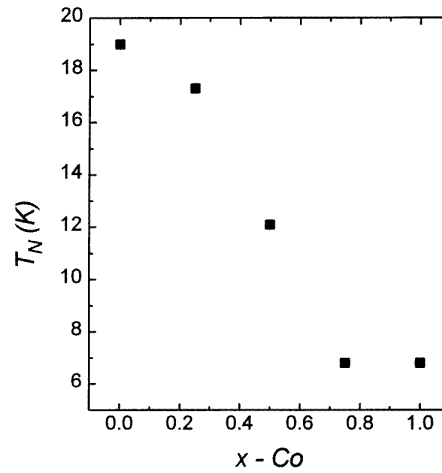
Correct analysis of the changes of the Néel temperature in the borocarbides with substitution and under pressure requires detailed knowledge of (anisotropic) RKKY interactions, and should be based on precise band structure calculations. We will try to obtain some qualitative insight into the problem on the basis of the simple isotropic model. For an isostructural series of metallic compounds, where the rare earth magnetic moments order via the (isotropic) RKKY interactions and in the absence of the crystalline electric field effects, the magnetic transition temperature  $T_m$  should scale as

$$T_m \propto N(E_F) \mathbf{J}^2 (g_J - 1)^2 \mathcal{J}(\mathcal{J} + 1) \quad (1)$$

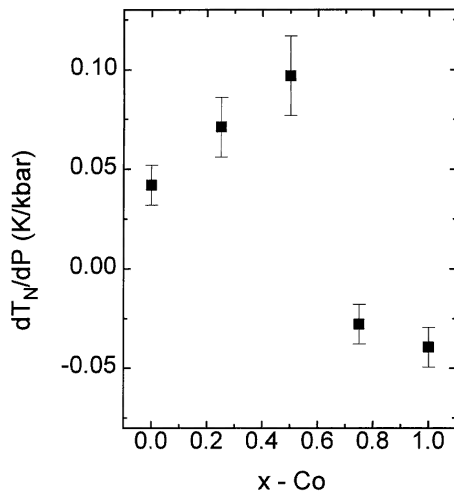
where  $\mathbf{J}$  is the (isotropic) conduction electron—rare earth exchange coupling parameter,  $N(E_F)$  is the DOS at the Fermi level,  $g_J$  is the Landé  $g$ -factor and  $\mathcal{J}$  is the total angular momentum of the rare earth atom [24]. In the case of Co substitution the main factor influencing  $T_m$  at ambient pressure is the modification induced in  $N(E_F)$ . As mentioned earlier, the variation of the lattice parameters (or the unit cell volume) from  $x = 0$  to  $x = 1$  is very small and it is of the same order of magnitude as the one induced by pressures between 15 and 25 kbar. Therefore, we do not expect noticeable modifications in the exchange coupling parameter with Co substitution. Although the results of the band structure calculations for the  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  series are not available in the literature,



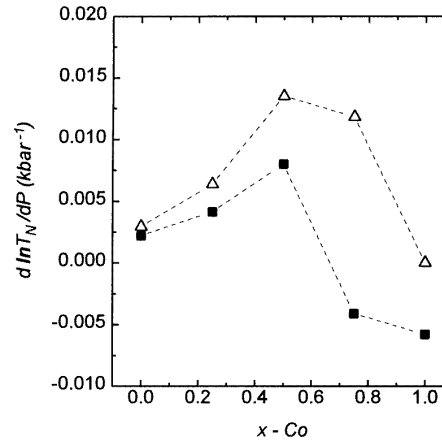
**Figure 3.** The low-temperature part of the resistance measurements of  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$ ,  $x = 0, 0.5$  and  $1.0$ .



**Figure 4.** The Néel temperature against Co concentration in the  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  series.



**Figure 5.** Pressure derivatives of the Néel temperature in the  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  series.



**Figure 6.** Relative changes of  $T_N$  under pressure,  $d \ln T_N/dP$ , against  $x$ :  $\blacksquare$ , experimental results;  $\triangle$ , results obtained from experimental  $T_N(x)$  dependence using (4).

it has been suggested that the band structure and the DOS are not significantly different for the different R in  $\text{RNi}_2\text{B}_2\text{C}$  [8]. The changes in the DOS at the Fermi level with Co substitution in  $\text{RNi}_2\text{B}_2\text{C}$  could be understood on the basis of the results of the band structure calculations [8] for  $\text{LuNi}_2\text{B}_2\text{C}$  and  $\text{LuCo}_2\text{B}_2\text{C}$ . While for  $\text{LuNi}_2\text{B}_2\text{C}$  the Fermi level is located near the maximum of the Ni(3d) peak in the DOS, for  $\text{LuCo}_2\text{B}_2\text{C}$ , due to the different band filling, the Fermi level is shifted about 0.8 eV to the low-energy side and

it is located on the shoulder of another peak in DOS in the vicinity of the local minimum between these peaks. Similar behaviour is obtained from cluster electronic calculations of Ni substituted in  $Y(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  compounds [25]. These results imply that the DOS at the Fermi level initially decreases with Co substitution, reaches some minimum value and starts to increase near  $x = 1$ . According to (1),  $T_N(x)$  is supposed to ‘trace’ the shape of the energy dependence of the DOS. This picture is consistent with the experimental  $T_N(x)$  dependence (figure 4).

In a simple mode, for fixed Co concentration, pressure causes an increase of the conduction electron density, which means a shift of the Fermi level to higher energies. In addition, pressure may cause modifications in the exchange coupling parameter. However, based on the data of the pressure dependence of the superconducting transition temperature for different  $\text{RNi}_2\text{B}_2\text{C}$ , the effect of pressure on the magnetic interactions between R ions was argued to be insignificant [12]. Moreover, the fact that the rapid changes of both  $T_N$  and  $dT_N/dP$  are observed in the same range of Co concentrations suggests the common origin of these changes, i.e. that the behaviour of  $N(E_F)$  under pressure gives the major contribution to  $dT_N/dP$ .

Neglecting in (1) the effect of pressure and the changes of the unit cell volume with Co substitution on the exchange coupling parameter  $\mathbf{J}$ , we will discuss the variation of  $T_N$  in our measurements. The differentiation of (1) will give for Co substitution at ambient pressure

$$d \ln T_N/dx = d \ln N(E_F)/dx = -1/(Z - x) d \ln N(E_F)/d \ln n \quad (2)$$

where  $n = N/V$  is the density of the conduction electrons;  $Z = 10$  is the (formal) number of valence electrons for Ni.

For the changes under pressure we will obtain, also by the differentiation of (1), now for a fixed Co concentration

$$d \ln T_N/dP = d \ln N(E_F)/dP = -(d \ln V/dP) d \ln N(E_F)/d \ln n. \quad (3)$$

Comparison of (2) and (3) gives

$$d \ln T_N/dP = (d \ln T_N/dx)(d \ln V/dP)(Z - x). \quad (4)$$

Figure 6 shows experimental values of  $d \ln T_N/dP$  with the values calculated with (4) using the data for  $T_N(x)$  and bulk modulus for  $\text{YNi}_2\text{B}_2\text{C}$ . Good qualitative agreement between the two sets of data in figure 6 supports the hypothesis that in both cases, with Co substitution and under pressure, the change of the DOS at the Fermi level is the determinant factor for the changes of the magnetic ordering temperature.

*In conclusion*, the pressure dependence of the magnetic ordering temperature was measured for the  $\text{Gd}(\text{Ni}_{1-x}\text{Co}_x)_2\text{B}_2\text{C}$  series. The changes of  $T_N$  both with Co substitution and under pressure could be explained by changes of  $N(E_F)$  due to the different band filling (substitution) or increase of the conduction electron density (pressure) as a major factor.

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