# $YNi_2B_2C$: possible anisotropic pressure dependence of the superconducting transition temperature



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# $YNi_2B_2C$: possible anisotropic pressure dependence of the superconducting transition temperature

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**Abstract.** The effect of chemical pressure on the superconductivity of the $(Y_{1-x}La_x)Ni_2B_2C$ system was studied. The results give an indication that the experimentally observed hydrostatic pressure for $YNi_2B_2C$ is possibly a consequence of the *balance* between *positive* in-plane and *negative* out-of-plane uniaxial pressure derivatives. It is suggested that *c*-uniaxial pressure is detrimental for superconductivity in this system.

## 1. Introduction

For several years the family of quaternary borocarbides $RNi_2B_2C$ (R—rare earth) [1–3] attracts a great deal of attention. The $RNi_2B_2C$ family has layered tetragonal crystallographic structure of the $ThCr_2Si_2$ type with alternating R–C and $(Ni–B)_2$ sheets, while band structure calculations [4–7], measurements of anisotropy of the superconducting critical field [8, 9] and normal state resistivity [10] strongly suggest a three-dimensional nature of superconductivity in the borocarbides. For pure $RNi_2B_2C$ compounds with R heavy rare earth both superconducting and magnetic ordering temperatures approximately scale with the de Gennes factor, while this scaling is seriously violated for R light rare earth. The absence of superconductivity in $PrNi_2B_2C$, $NdNi_2B_2C$ and $SmNi_2B_2C$ was argued [11] to be related to the difference in exchange polarization of conduction electrons in compounds with light and heavy R. In the present work we attempt to link the structural changes with the superconducting properties in the $(Y_{1-x}La_x)Ni_2B_2C$ series of the borocarbides, where non-magnetic isoelectronic La doping is performed at the rare earth site, by analysing the results together with the literature data on pressure effect on $YNi_2B_2C$.

## 2. Experiment

Polycrystalline $(Y_{1-x}La_x)Ni_2B_2C$ samples were prepared by a conventional arc-melting of stoichiometric amounts of pure elements in Ar atmosphere. To ensure homogeneous samples,

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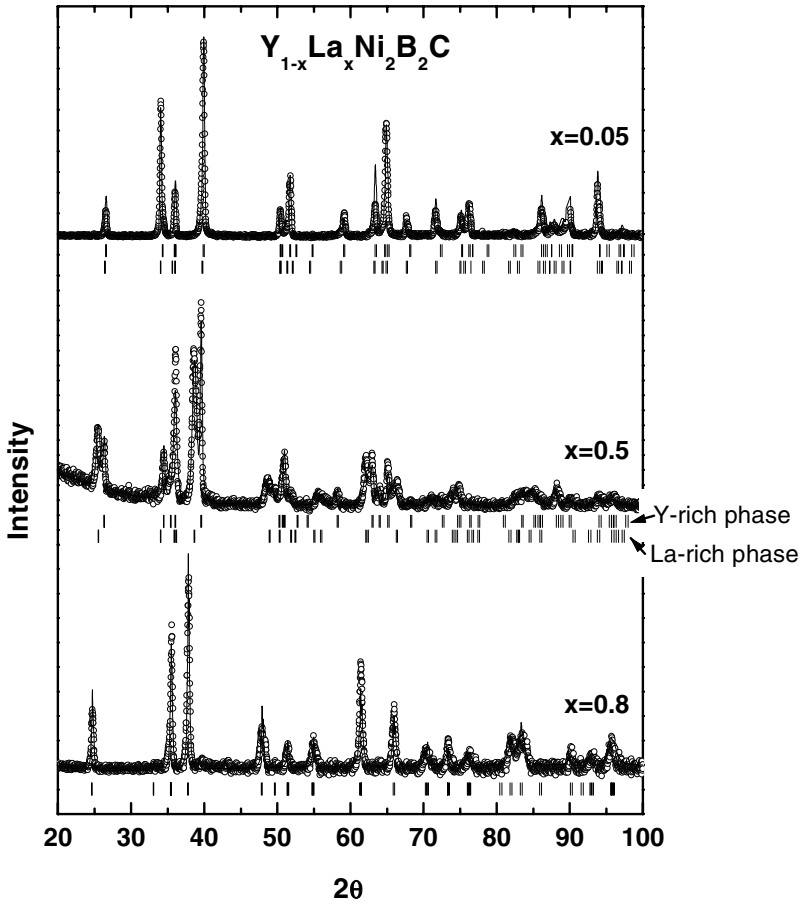
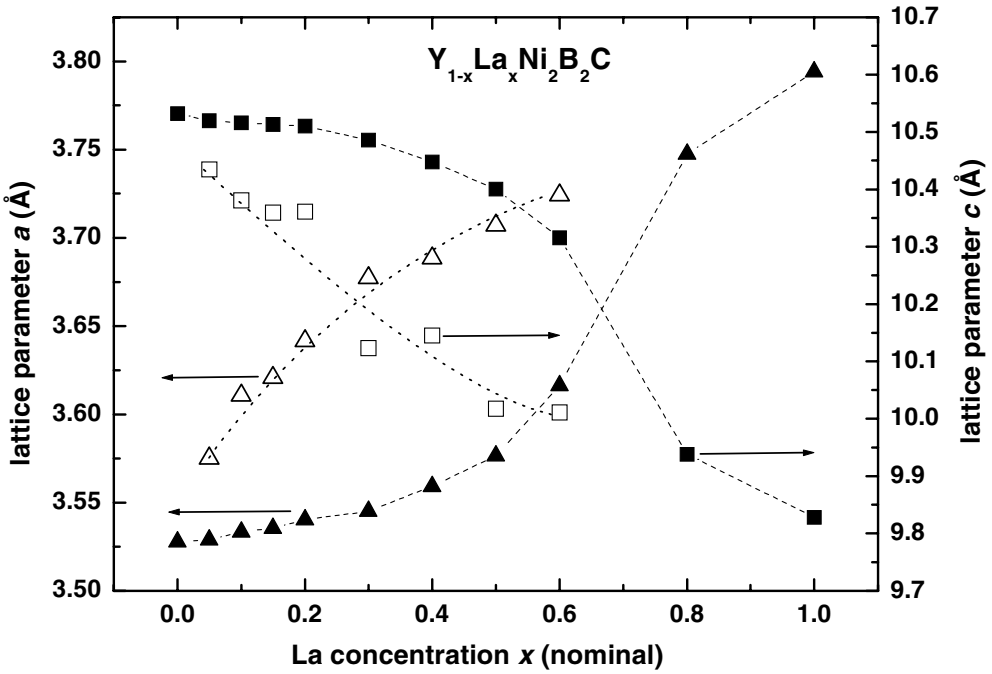



(a)

**Figure 1.** (a) Cu K$\alpha$ x-ray diffraction pattern of the samples $(Y_{1-x}La_x)Ni_2B_2C$ with $x = 0.05$, 0.5 and 0.8. The data with $x = 0.5$ can be well analysed with two patterns with $I4/mmm$ space group symmetry. The upper vertical lines are the Bragg peaks corresponding to the Y-rich phase and the lower ones to the La-rich phase. (b) Lattice parameters of the main Y-rich phase (closed symbols) and the secondary phase (open symbols) of the $(Y_{1-x}La_x)Ni_2B_2C$ series as a function of the nominal La doping.

they were turned over and re-melted four times. Then, the samples were wrapped in tantalum foil, vacuum sealed in a quartz tube and annealed at 1050 °C for two days. This cycle was repeated twice. The room temperature x-ray diffraction measurements were performed on powdered samples with a Rigaku Miniflex diffractometer using Cu K$\alpha$ radiation. Rietveld analysis was used to obtain the lattice parameters of the samples. Magnetization measurements were done in a Quantum Design superconducting quantum interference device (SQUID) magnetometer in a temperature range between 5 and 20 K. For the samples with nominal La concentration of $x = 0.5$ and 0.6, AC susceptibility ($\chi'_{AC}$) measurements were performed between 1.8 and 15 K in a modulation field $h_{ac} \approx 1$ G using a homemade susceptometer. $T_c$ was defined as the maximum slope of $\partial\chi_{DC}/\partial T$ or $\partial\chi'_{AC}/\partial T$, where $\chi_{DC} = M/H$, and the width at half height of their respective curves (assumed as Gaussians) were taken as a rough approximation of the error in $T_c$.




(b)

**Figure 1.** (Continued)

## 3. Results

The x-ray analysis of $(Y_{1-x}La_x)Ni_2B_2C$ confirms a nearly single phase with good crystallinity only for compositions close to the Y and La ends (figure 1(a)). For nominal concentration with $0.05 \leqslant x \leqslant 0.7$ (miscibility gap) two phases with $LuNi_2B_2C$ type of structure were found: for $0.05 \leqslant x \leqslant 0.3$ two Y-rich phases and for $0.3 \leqslant x \leqslant 0.7$ a Y-rich phase and a La-rich phase, were needed to analyse the x-ray patterns. Figure 1(a) shows the results of the Rietveld analysis assuming the presence of the above-mentioned phases for the samples with nominal concentrations $x = 0.05$, 0.5 and 0.8.

In figure 1(b). the lattice parameters of both the main Y-rich phase (closed symbols) and the secondary phase (open symbols) are plotted as a function of the nominal La concentration. For the secondary phase the lattice parameter $a$ ($c$) increases (decreases) with the La concentration (open symbols in figure 1(b)), giving a clear indication that this phase becomes more La rich as the nominal lanthanum concentration increases. Similar miscibility gap has been also reported for $(Ho_{1-x}La_x)Ni_2B_2C$ [12].

The effect of La substitution on the superconducting transition can be seen in figure 2; there is a decrease of $T_c$ with a broadening of the superconducting transition and a reduction of the superconducting fraction. Taking into account that the $LaNi_2B_2C$ is not superconducting, it is natural to suppose that superconductivity of $YNi_2B_2C$ is suppressed by substitution of Y by La. Therefore, we attribute the superconducting signal observed by magnetization measurements in the $0.05 \leqslant x \leqslant 0.6$ region of nominal La concentrations to the Y-rich phase (figure 2).

The correlation between $T_c$ and $x$ is not easily made because the nominal concentration does not correspond to the real concentration in the main Y-rich phase. If we assume that the

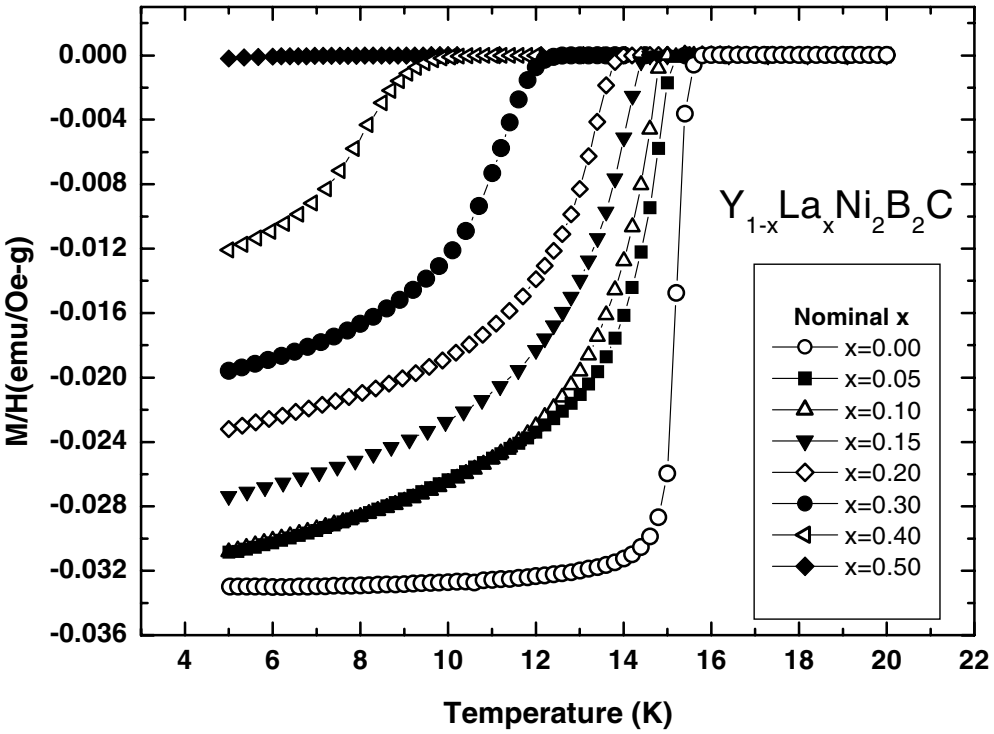



**Figure 2.** Magnetization curves for $(Y_{1-x}La_x)Ni_2B_2C$ for different nominal concentrations of La.

lattice parameters vary linearly, going from pure $YNi_2B_2C$ to pure $LaNi_2B_2C$, as the La doping increases, we can make a rough estimation of the La concentration in the samples under study and the results are plotted in figure 3. As we will see below, our final result does not change significantly if we use these estimated values $x$ of La or the nominal ones. Therefore, we use the estimated $x$ values in figure 4 to plot the variation of $T_c$ with the La concentration. The data contained in figures 3 and 4 will be used in our analysis.

Superconducting transition temperature decreases monotonically with La concentration, superconductivity being completely suppressed at about the estimated La concentration $x \approx 0.45$ (figure 4). $T_c$ decreases linearly for low La concentration (for estimated concentration $x \leqslant 0.2$); for higher La concentration this linearity is broken.

## 4. Analysis

To analyse our results the following assumptions were made: (1) the effect of disorder on $T_c$ is negligible for low La concentrations, (2) the effects of chemical and physical pressure on $T_c$ are equivalent [13].

For *La substitution* (considering the linear part):

$$dT_c/dx = 2\partial T_c/\partial a\, da/dx + \partial T_c/\partial c\, dc/dx. \tag{1}$$

For *hydrostatic pressure*:

$$dT_c/dP = 2\partial T_c/\partial a\, da/dP + \partial T_c/\partial c\, dc/dP. \tag{2}$$

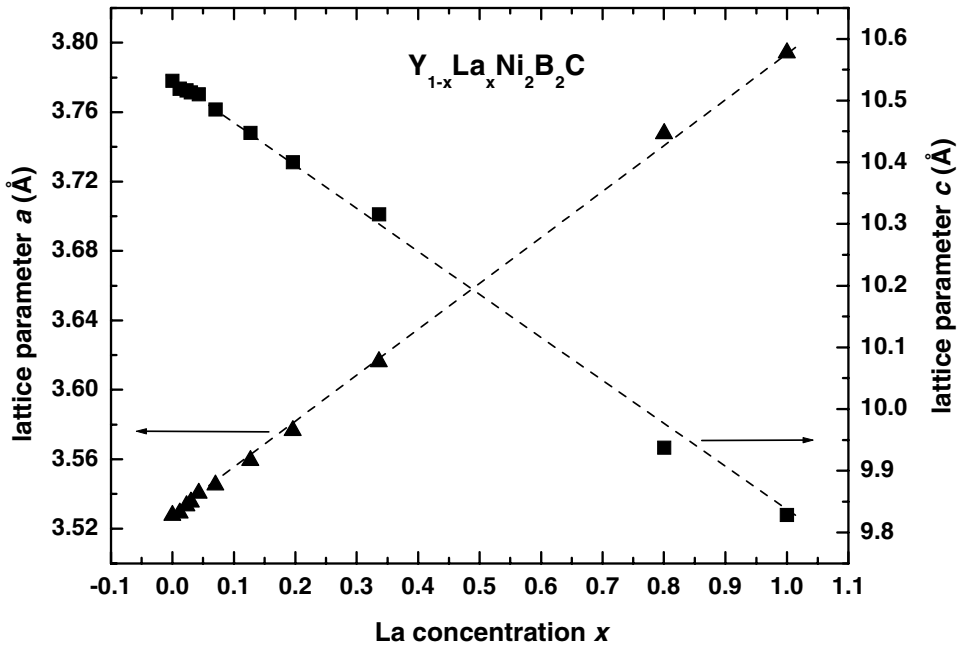



**Figure 3.** Lattice parameters for the main Y-rich phase of the $(Y_{1-x}La_x)Ni_2B_2C$ series as a function of the estimated La concentration (assuming a linear variation of the lattice parameters going from $YNi_2B_2C$ to $LaNi_2B_2C$).

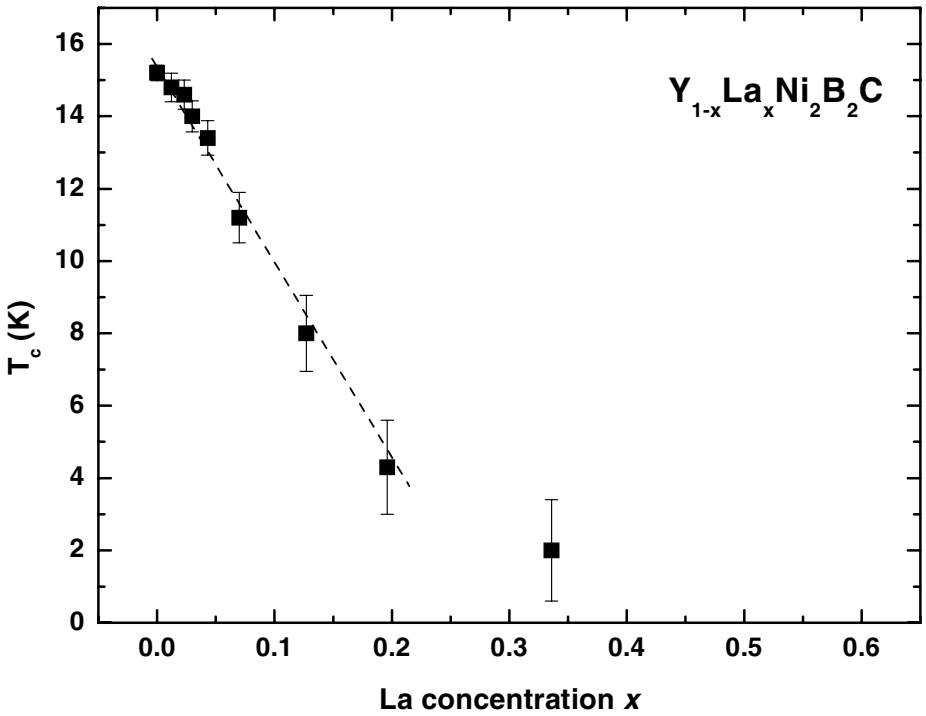



**Figure 4.** Superconducting transition temperatures of the $(Y_{1-x}La_x)Ni_2B_2C$ series as a function of estimated La concentrations.

From results shown in figures 3 and 4, $dT_c/dx$, $da/dx$ and $dc/dx$ are estimated (for the linear part $x \leqslant 0.2$) as:

$$dT_c/dx \approx -54.29 \text{ K}, da/dx \approx 0.2504 \text{ Å and } dc/dx \approx -0.6678 \text{ Å}.$$

Pressure derivatives of the superconducting transition temperature $dT_c/dP$ for $YNi_2B_2C$ were measured by several groups. The reported results are quite scattered:

$-0.58 \times 10^{-2}$ K kbar$^{-1}$[14]; $-0.9 \times 10^{-2}$ K kbar$^{-1}$[15];

$0.32 \times 10^{-2}$ K kbar$^{-1}$ $- 0.313 \times 10^{-2} P$ K kbar$^{-2}$[16].

Due to the differences in the available literature data, the value $dT_c/dP = -0.6 \times 10^{-2}$ K kbar$^{-1}$ will be used in the following analysis, although the results will be qualitatively the same if other numbers from the above are used. The compressibility of $YNi_2B_2C$ was measured in [17]:

$$d \ln a/dP = -0.22 \times 10^{-3} \text{ kbar}^{-1}, d \ln c/dP = -0.40 \times 10^{-3} \text{ kbar}^{-1}.$$

Solving together (1) and (2) we obtain $\partial T_c/\partial a$ and $\partial T_c/\partial c$:

$$\partial T_c/\partial a \approx -71.56 \text{ K Å}^{-1}, \partial T_c/\partial c \approx 27.72 \text{ K Å}^{-1}$$

or written in another way:

$$\partial T_c/\partial P_a \approx 0.055 \text{ K kbar}^{-1}, \partial T_c/\partial P_c \approx -0.116 \text{ K kbar}^{-1}.$$

We want to point out that if we use the nominal La concentration and the values of $a$, $c$ and $T_c$ in the range $0.0 \leqslant$ nominal $x \leqslant 0.3$ (linear part in figure 1(b)) for our calculations we get $\partial T_c/\partial P_a \approx 0.046$ K kbar$^{-1}$ and $\partial T_c/\partial P_c \approx -0.098$ K kbar$^{-1}$, which are close to the previous calculated values. This behaviour can be understood taking into account that the variations $da/dx$ and $dc/dx$ are counterbalanced by $dT_c/dx$ in equation (1). Then, it may be more realistic to take an average of the two values for the variation of $T_c$ with the uniaxial pressures:

$\partial T_c/\partial P_a \approx 0.0505 \pm 0.005$ K kbar$^{-1}$, $\partial T_c/\partial P_c \approx -0.107 \pm 0.01$ K kbar$^{-1}$[18].

The obtained values suggest that the small experimental hydrostatic pressure dependences $dT_c/dP$ for $YNi_2B_2C$ are the result of the *balance* between *positive* in-plane and *negative* out-of-plane uniaxial pressure derivatives. These results may indicate the anisotropic effects of the crystalline structure on the otherwise three-dimensional superconductivity in $YNi_2B_2C$. The above estimations are qualitatively in agreement with the band structure calculations of Mattheiss *et al* [19], where it was shown that superconductivity in the quaternary borocarbides is particularly sensitive to the $NiB_4$ tetrahedral geometry (i.e. for example to $c/a$ ratio) (figure 5) and that favourable superconducting properties are expected for the compounds where the $NiB_4$ tetrahedra are nearly 'ideal' ($LuNi_2B_2C$, $YNi_2B_2C$). Since for all studied quaternary borocarbides the tetrahedral bond angles $\alpha < \alpha_{ideal}$, the above mentioned work anticipated that the uniaxial pressure *along the c-axis* would be detrimental for superconductivity, which is consistent with the result of present study.

Another point to mention is related to the absence of superconductivity in the light rare-earth quaternary borocarbides. As mentioned above, the first attempt to explain this lack of superconductivity was made by Mattheiss *et al* [19], emphasizing the relevance of the 'perfect' tetrahedral angle for the occurrence of superconductivity. If we assume that that $z$-parameter of boron in the unit cell is constant through the whole series (the actual change [20] is about 3.6% from $LaNi_2B_2C$ to $LuNi_2B_2C$), the tetrahedral bond angles will be determined by the $c/a$-ratio. Taking the $T_c$–$c/a$ graph for the $(Y_{1-x}La_x)Ni_2B_2C$ series (figure 6) as a very rough approximation of the behaviour in the $RNi_2B_2C$ series, we will expect that the changes in

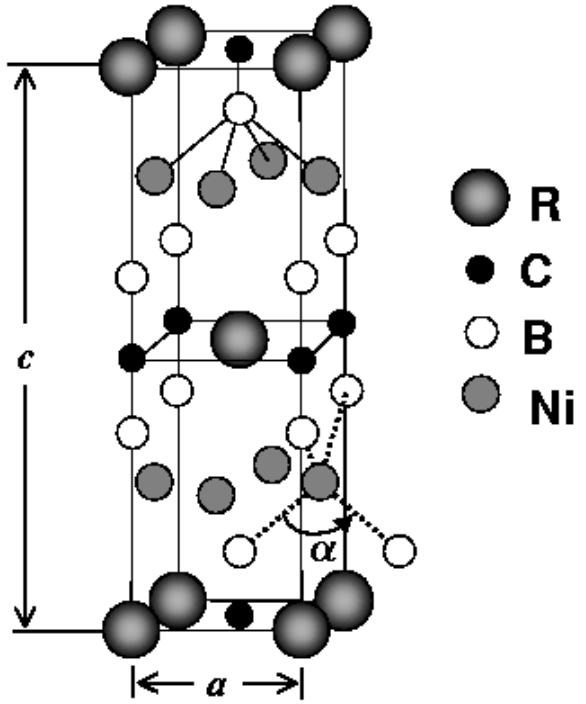



**Figure 5.** $NiB_4$ tetrahedral geometry in the $LuNi_2B_2C$ structure.

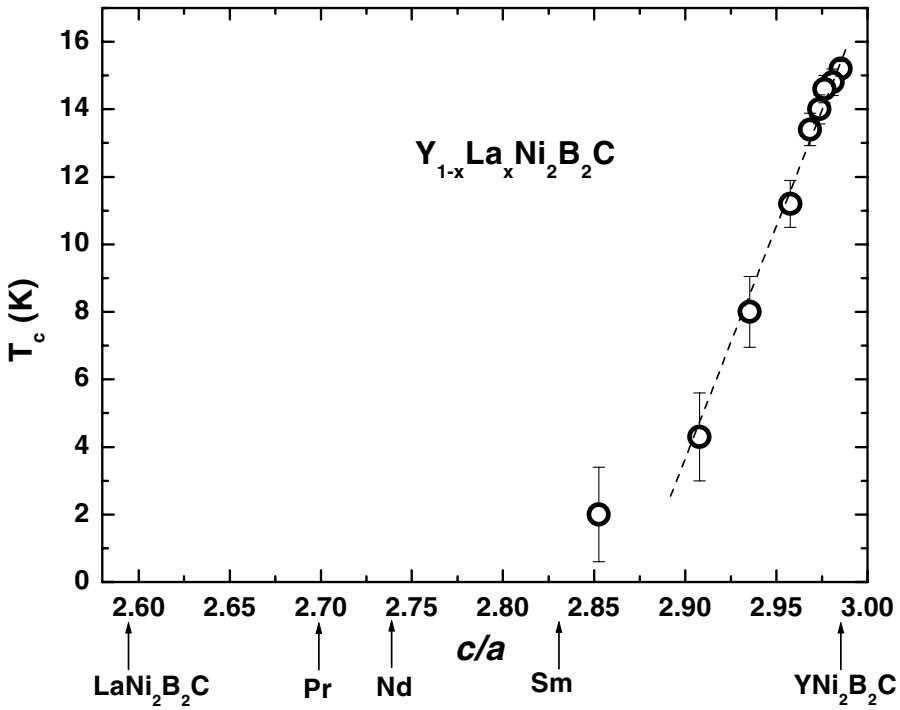



**Figure 6.** Variation of $T_c$ with the ratio $c/a$ for the $(Y_{1-x}La_x)Ni_2B_2C$ series.

the lattice parameters with the rare earth may have significant detrimental contribution to the superconductivity for the light rare earth borocarbides and should be included in the analysis of the ground state properties of these compounds.

The study of the universality of $T_c$–$c/a$ behaviour in other non-magnetic rare earth substitutions, e.g. $(Lu_{1-x}La_x)Ni_2B_2C$ [21], as well as the equivalence of physical and chemical pressure for quaternary borocarbides should be very interesting.

## 5. Conclusion

In conclusion, a decrease of $T_c$ with the substitution of La by Y was observed in $(Y_{1-x}La_x)Ni_2B_2C$. The simultaneous analysis of chemical and physical pressure dependences suggests that small pressure derivatives found in $YNi_2B_2C$ are the result of the *balance* between *positive* in-plane and *negative* out-of-plane uniaxial pressure responses

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