

Pressure effects in PrT_2B_2C (T = Co, Ni, Pt): Applied and chemical pressure

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High pressure electrical resistivity ρ_{ab} measurements on intermetallic Pr(Co, Ni, Pt)₂B₂C compounds were performed down to 2 K. At ambient pressure the $\rho_{ab}(T)$ curves for the nonsuperconducting Pr(Co, Ni)₂B₂C compounds exhibit magnetic correlations at about 10 and 4 K, respectively. At low temperatures, PrCo₂B₂C shows a large spin-dependent electron scattering in comparison to PrNi₂B₂C. Under applied pressure, the magnetic scattering tends to be suppressed more effectively in PrCo₂B₂C than in PrNi₂B₂C. The low-temperature behavior of $\rho_{ab}(T,P)$ for PrNi₂B₂C and PrCo₂B₂C suggests a spin fluctuation mechanism. On the other hand, the PrPt₂B₂C compound shows superconductivity at about 6 K and under pressure its superconducting transition temperature tends to be degraded at a rate $dT_c/dP = -0.34$ K/GPa, as expected in compounds with transition metals. The experimental results in Co-, Ni-, and Pt-based compounds are analyzed from the point of view of the external and chemical internal pressure effects.

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1 Introduction The phenomenon of superconductivity appearing in compounds having magnetic elements has received noticeable attention during the last three decades due to the great variety of exotic electronic and magnetic correlations it involves. Particularly interesting for this topic has been the discovery of the quaternary intermetallic compounds RNi_2B_2C (R = rare earths, Y, Sc, Th) [1, 2], where coexistence of antiferromagnetism and superconductivity has been observed as for example in R = (Tm, Er, Ho,Dy, Lu) [2–7]. As far as we know, several RT_2B_2C intermetallic compounds, with different rare earths (R) and transition metal (T) combinations, have been synthesized [3, 6, 8–14] and most of them, such as HoNi₂B₂C [15], show superconductivity in spite of the presence of the rare earth magnetic element. From the theoretical [16] and experimental [17] points of view, a conventional electron-phonon mechanism has been claimed to be responsible for superconductivity in these materials. The influence of the transition metal magnetism on the magnetic properties of these compounds seems to be of minor importance compared to that of the rare earth ions, whose magnetic moments apparently impose the magnetic ordering at all. Thus, in some borocarbides with 3d transition elements such as Ni and Co, neutron-diffraction measurements [18] and electronic transport measurements [11, 19] have revealed that no significant magnetic moment develops in the T sites. Local structure studies at the Ni site, using Mössbauer spectroscopy on ⁵⁷Fe-doped (1 at%) samples, also support this fact [20]. Interestingly, the T elements play an indirect role in the magnetism of magnetic RT₂B₂C systems through the spatially dependent indirect RKKY exchange interactions [7, 21] that govern the magnetic ordering in these compounds. On the other hand, the electronic influence of the T elements on the superconductivity of RT₂B₂C is more relevant than that for the R, B, and C elements. This is particularly true for the Ni-based borocarbide superconductors, where the density of state at the Fermi level is mainly due to the Ni 3d bands [22]. On the contrary, in a



comparative study of the structure and superconducting properties of RNi₂B₂C, Loureiro et al. [22] showed that the superconducting state is more strongly affected by the magnetism of the R ion than by the R-ion size, at least for R between Dy and Tm. However, the role of the magnetism and ion size of T elements in the superconductivity of RT₂B₂C when magnetic R ions are present is not clear yet. In this work we study a particularly interesting case: the PrT₂B₂C compounds with T = Ni, Co, and Pt, which have revealed many peculiarities: PrNi2B2C and PrCo2B2C do not superconduct as measured down to 0.3 K [23]; however, PrPt₂B₂C does superconduct at 6 K, even in the magnetic Pr⁺³-ion presence [3, 24]. Noticeably, PrPt₂B₂C does not show any magnetic ordering at low temperatures [24] but, in contrast, Pr-Ni and Pr-Co based borocarbides develop antiferromagnetic ordering at about 4 and 8.5 K, respectively [25, 26]. Recently, magnetoresistance and specific heat studies in Pr(Co, Pt)₂B₂C [26, 27] have pointed out that a spin fluctuation mechanism is involved in the electronic behavior of these two compounds. However, although evidence for spin fluctuations can be deduced from certain features in the electronic transport measurements, the interpretation of those properties is not so clear. High-pressure experiments in spin fluctuators such as RCo₂ [28], CeNi₅ [29], and UPt₃ [30] have proven to be a useful tool in order to make clear if a spin fluctuation mechanism is occurring in such systems. The aim of this paper is to enlighten the influence of the chemical and external applied pressure on the superconducting state and magnetic scattering at low temperature for the three Pr-based borocarbides: Pr(Ni, Co, Pt)₂B₂C. We analyzed the changes of the resistivity as a function of pressure and temperature. We assume that interactions between itinerant electrons play an important role in the low temperature resistivity characteristic, and those can be modified by applied external or internal chemical pressure.

2 Experimental details Three compounds were prepared: samples of PrCo₂B₂C, PrNi₂B₂C, and PrPt₂B₂C. The single crystals were grown by a cold copper crucible method as described by Durán et al. [26]. All samples were characterized by X-ray diffraction using a Bruker P4 diffractometer, with monochromatized Mo-Ka radiation. The cell parameters were: a = 3.6156(1) Å and c = 10.3507(6) Å for PrCo₂B₂C, a = 3.6996(2) Å and c = 9.9885(8)Å for PrNi₂B₂C and a = 3.8373(1)Å and c = 10.7610(9)Å for PrPt₂B₂C samples. Resistivity measurements in the a-b plane were performed by the four-probe technique using gold wires of 10-µm diameter as electrical contacts. Pressure experiments were performed by using a micro-cryogenic diamond anvil cell (MCDAC, piston-cylinder type Be-Cu cell) consisting of two diamonds, each of 0.5-mm culet size. A Cu-Be gasket was preindented and a 150-µm-diameter hole was drilled at the center. The samples used have dimensions of about $80 \times 15 \times 40 \,\mu\text{m}^3$ and were placed in the gasket hole. The transmitting pressure medium was MgO powder. The metallic gasket was electrically insulated by pressing over

it Al₂O₃ powder of 1- μ m grain size. As the MCDAC pressure increases, the wires used to measure the electrical resistance may be cut off at the edge of the diamonds because of the diamond indentation. To reduce this problem, we used a thin aluminium foil placed under the four gold wires; with this setup, we frequently reached high quasi-hydrostatic pressures in the range of about 6 GPa. Also, in order to prevent motion of the sample and of the electrical leads at the initial compressing, a thin mylar film was placed over them. Additional pressure experiments in polycrystalline Pr(Ni, Pt)₂B₂C compounds up to about 21 GPa were made using a sintered diamond Bridgman anvil apparatus with a pyrophyllite gasket and two steatite disks as the pressure medium. For determination of the pressures, a Pb manometer was used.

3 Results and discussion The RNi₂B₂C compounds crystallize in the tetragonal body-centered structure (space group I4/mmm) and, when the rare earth atom radius is increased (R goes from La to Lu), the *c* parameter becomes larger whereas the *a* parameter decreases [31, 18]. This structural behavior can be accounted for by the rigidity of the B–C and Ni–B bonds and the variable tetrahedral angle in the NiB₄ unit. Distortions of this tetrahedral unit are claimed to be a decisive parameter for T_c in non-magnetic or antiferromagnetic RNi₂B₂C and RNiBC compounds [32]. In the case when the size of the transition element is increased, maintaining the same rare earth element, the structural behavior of the unit cell seems to be slightly different. Figure 1 displays the T–T shortest length between 3d ions,



Figure 1 (online color at: www.pss-a.com) Variation of the T–T shortest length between 3d ions for the transition metal T and PrT_2B_2C compounds, with T = (Co, Ni, Pt). It also shows the behavior of the cell parameters as a function of the T size.

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Figure 2 (online color at: www.pss-a.com) Normalized resistivity at 295 K ($\rho_{ab}(T)/\rho_{ab}(295 \text{ K})$) of PrNi₂B₂C, PrCo₂B₂C, and PrPt₂B₂C single crystals at ambient pressure. The three systems present metallic conductivity from room temperature down to 25 K. Inset shows the low-temperature variation of the normalized resistivity.

particularly in the $Pr(Ni, Co, Pt)_2B_2C$ compounds and that for the metal T. Also, it shows the behavior of the *a* and *c* parameters for each compound. We note that increasing the ionic radius T size causes an increase of the *c* parameter and an anomalous behavior of the *a* parameter for the $PrCo_2B_2C$ compound. At first glance and, according to the figure, this anomaly is correlated with the variation of the T–T shortest bond in the framework of the PrT_2B_2C structure, and not with the ionic radius size of the T element.

Figure 2 shows the normalized electrical resistivity in the a-b plane as a function of temperature for PrNi₂B₂C, PrCo₂B₂C, and PrPt₂B₂C single crystals at ambient pressure. The three compounds present metallic characteristics. At low temperature the Ni- and Co-based compounds show notable similarities, but are not superconductors, whereas PrPt₂B₂C has a sharp superconducting temperature at about 6 K. The residual resistivity, residual resistivity ratio (RRR), quadratic coefficient, superconducting transition temperature, Sommerfeld coefficient, and change rate of T_c under pressure parameters for the three systems are listed in Table 1. The residual resistivity ρ_0 of all three is sample dependent, varying between 5 and 25 $\mu\Omega$ cm. The residual resistivity ratios RRRs for the first two compounds are 9 and 33, respectively, whereas for PrPt₂B₂C RRR is 5.5. These

values are similar to those reported in a previous work [26]. The main panel of Fig. 2 shows some interesting characteristics: Ni- and Co-based compounds present a notable positive curvature from about 150 to 50 K, whereas the Pt compound presents a wide bump from about 250 to 20 K. These notable differences may signal a clearly distinctive influence of the crystalline field at high temperatures. A gradual but pronounced drop in resistivity disturbs the linear variation to about 8 and 20 K for PrNi₂B₂C and PrCo₂B₂C, respectively. Such resistivity behavior at relatively low temperature is typical for magnetic elements of the RNi₂B₂C series, and it has been associated with a decrease of the magnetic scattering of the conduction electrons by rare earth ions (see Ref. [33] and references therein). However, according to the result that will be presented, it is possible that another mechanism involving conduction electrons could also develop at low temperature. The case for PrPt₂B₂C is quite different; after following an upward curvature, it becomes superconducting at about 6K. Magnetic and heat capacity measurements in this compound [24] have revealed a non-magnetic ground state for Pr ions due to crystalline electric field (CEF) effects, which is claimed to be the reason for superconductivity. The inset of Fig. 2 shows the resistivity behavior from 60 to 2 K for the three compounds at ambient pressure. At first glance, increasing the transition metal radius corresponds to a major resistivity drop at low temperature.

The $\rho_{ab}(T)$ curve for PrCo₂B₂C from about 2 to 8 K shows a clear T^2 -law dependence with a quadratic coefficient A equal to 0.08 $\mu\Omega$ cm/K², see Fig. 3. This low temperature resistivity behavior is similar to that observed in heavy fermion systems, as for example YbNi₂B₂C and UPt₃ compounds [34, 35], and could be attributed to spin fluctuations [27, 36]. Thus, in a similar compound but simpler, RCo₂, a T^2 dependence has been found at low temperatures, which is due to spin fluctuation characteristics [28]. The fact that the magnitude of the quadratic coefficient A of $\rho(T)$ for PrCo₂B₂C is of the order of that for RCo₂ (Massalami et al. [37] found this coefficient as big as three orders of magnitude but in polycrystalline PrCo₂B₂C) suggests that spin fluctuations could be the responsible mechanism for the low temperature $\rho(T)$ behavior in this compound. Using the universal relation for heavy fermion compounds, $A/\gamma^2 = 1.0 \times 10^{-5} \mu \Omega \text{cm}(\text{mol K})^2/\text{mJ}^2$ [38], the resulting Sommerfeld coefficient is $\gamma = 89.4 \text{ mJ/mol K}^2$, which is a low value compared with that for PrNi₂B₂C (211 mJ/ mol K² [26] by specific heat measurements), but an enhanced

Table 1 ρ_0 , residual resistivity; RRR, residual resistivity ratio; A, quadratic coefficient; γ , Sommerfeld coefficient at ambient pressure for the Pr(Ni,Co,Pt)₂B₂C compounds.

compound	a (Å)	<i>c</i> (Å)	$ ho_0$ ($\mu\Omega$ cm)	RRR	$A \\ (\mu\Omega \text{cm/K}^2)$	<i>T</i> _c (K)	γ (mJ/molK ²)	d <i>T</i> _c /d <i>P</i> (K/GPa)
PrNi ₂ B ₂ C	3.6996(2)	9.9885(8)	3.4	9.0			211.0	
PrCo ₂ B ₂ C	3.6156(1)	10.3507(6)	1.67	33.0	0.08		89.4	
PrPt ₂ B ₂ C	3.8373(1)	10.7610(9)	25.0	5.5		6.0		-0.34





Figure 3 (online color at: www.pss-a.com) Linear fit to $\rho(T^2)$ from 2 to 8 K for PrCo₂B₂C. The low-temperature behavior of $\rho(T)$ follows a T^2 law with a quadratic coefficient $A = 0.08 \ \mu\Omega$ cm/K². Inset shows the $\rho(T)$ behavior at low temperatures.

value as compared to the normal metal Co and to other borocarbides such as YCo_2B_2C [11] and (Gd, Tb, Dy, Ho, Er, Tm)Ni₂B₂C [39], whose γ is about 17 mJ/mol K².

Figures 4 and 5 show the $\rho_{ab}(T)$ curves as a function of pressure for PrNi₂B₂C and PrCo₂B₂C single crystals, respectively. As we can see, these compounds reveal different pressure behaviors. The overall trend of $\rho_{ab}(T)$ for the Pr–Ni based compound does not change as the pressure increases up to 5.3 GPa. The linear behavior of $\rho_{ab}(T)$ (extending from about 100 K to ambient temperature) is attributed to electron–phonon scattering and, under the applied pressures, it shows a slope decreasing from 0.089 to 0.069 $\mu\Omega$ cm/K. According to the inset of Fig. 4, the smooth drop of $\rho_{ab}(T)$ at low temperature, which has been related to



Figure 4 (online color at: www.pss-a.com) The graph shows pressure effects on $\rho_{ab}(T)$ for a PrNi₂B₂C single crystal up to 5.3 GPa. Vertical line indicates the increasing pressure. Inset is a zoom of $\rho_{ab}(T, P)$ at low temperature.



Figure 5 (online color at: www.pss-a.com) In panel (a) there is presented $\rho_{ab}(T)$ for a PrCo₂B₂C single crystal under several pressures up to 4.4 GPa. Panel (b) shows a view of $\rho_{ab}(T)$ at low temperatures and for different pressure values.

the decrease of magnetic scattering [33], is reduced with applied pressures increasing up to 5.3 GPa. In the case of $PrCo_2B_2C$, as can be seen in panel (b) of Fig. 5, the pressure effects are stronger than in PrNi₂B₂C, mainly in the lowtemperature regime. From ambient pressure to about 1.7 GPa the high-temperature behavior of $\rho_{ab}(T)$, from 300 to about 75 K remains without appreciable changes and with almost a constant slope of $0.30 \,\mu\Omega \text{cm/K}$. In an opposite way to PrNi₂B₂C, the low-temperature curvature of $\rho_{ab}(T)$, which is also associated with magnetic correlations, tends strongly to be suppressed by pressure. This tendency has also been observed by Massalami et al. [37] by applying pressures up to about 1.2 GPa, the maximum pressure value they applied. Similar to their results, we also observed that the quadratic behavior of $\rho_{ab}(T)$ at low temperatures is maintained under 1.2 GPa. However, we found, by applying pressures higher than 1.7 GPa to this compound, a distinctive characteristic, namely the change of the low-temperature curvature from concave to convex and the complete disappearance of the resistivity drop above 2.9 GPa (see panel (b) of Fig. 5). Interestingly, at this pressure the T^2 behavior disappeared and instead there is the appearance of a type of plateau zone in $\rho_{ab}(T)$ which starts at about 15 K and extends down to 1.8 K, the lowest temperature available in our experiments. Increasing the pressure up to 4.4 GPa, this zone of constant resistivity is extended from 1.8 up to 20 K. At this pressure, the overall high-temperature behavior of $\rho_{ab}(T)$ remains almost with the same slope of about $0.30 \,\mu\Omega$ cm/K. The above experimental facts reveal that the electronic properties

of PrCo₂B₂C are more pressure sensitive than those of PrNi₂B₂C, mainly in the low-temperature regime. Interestingly, we note that the shape of $\rho_{ab}(T)$ for PrCo₂B₂C under pressure tends to be qualitatively similar to that for non-magnetic YCo₂B₂C at ambient pressure [11]. Thus, it seems that the effect of pressure in this compound is to suppress the magnetic correlations which cause the low temperature scattering behavior.

According to Fig. 5, a T^2 behavior can be fitted to the $\rho_{ab}(T)$ curve corresponding to ambient pressure only for a region of relatively low temperatures. Interestingly, such a fitting range is extended when the pressure does increase. As we already pointed out, at atmospheric pressure the fitting goes from 2 up to about 8 K with a quadratic coefficient A equal to 0.08 $\mu\Omega$ cm/K². At 1.7 GPa the interval extends from 2 to about 19.5 K, with a value of A decreased to $0.018\,\mu\Omega\text{cm/K}^2.$ Once the applied pressure reaches the value of 2.9 GPa, it was no longer possible to fit a quadratic function to the curvature of $\rho_{ab}(T)$ because of the plateaulike zone, also presented for the curves at 3.7 and 4.4 GPa. This $\rho_{ab}(T)$ behavior at low temperature and pressures is accounted for by the spin fluctuation scenario [40], which also takes into account the decreasing of the A parameter with the applied pressure. Additionally, relatively low magnetic fields decrease the A parameter in a linear form in $PrCo_2B_2C$ [27], which has been claimed to be due to quenching of the spin fluctuation. Similar results have been found in other systems such as for example Ce_{0.8}(Pr, Nd)Ni₅, indicating that spin fluctuations tend to be suppressed by both pressure and magnetic fields [41, 42]. The microscopic character of the state resulting from applying a magnetic field is completely different from that obtained with applying pressure; the first remains magnetic, whereas the second tends to be a real non-magnetic state; one where no microscopic magnetic moments still exist, but long-range order has been destroyed by the fluctuations. This is important because in the spin fluctuation model the state above $T_{\rm sf}$ (the temperature at which the spin fluctuation appears and which coincides with the temperature below which a T^2 law in resistivity is valid) is not the non-magnetic state (like in the Stoner model) but a magnetic state, where local moments still exist, but long-range order tends to be destroyed by the fluctuations. The collective modes described by the spin fluctuations can readily be excited at relatively low temperature, where the Stoner excitations are still very small but we assume that they can be suppressed by two factors: intense magnetic field and/or external applied pressure. Pressure increases the correlation that exists between f ions and promotes the itinerance of f electrons. As a result, the f-density of state near the Fermi level is lowered, modifying the electron structure and influencing thus the prevailing long-range order between the band electrons.

In order to know if $\rho(T)$ of PrNi₂B₂C could follow a similar behavior to that for its isomorph PrCo₂B₂C at higher pressures, we carried out measurements for a polycrystalline sample at several pressures up to 21.5 GPa. Interestingly, we

found that there is a marked tendency of $\rho(T)$ at low temperature to behave similarly to that for PrCo₂B₂C (see Fig. 6). At about 13.5 GPa there is a change from negative to positive curvature of $\rho(T)$ at temperatures lower than about 15 K. This curvature change was also found in the Pr-Co system but at about 1.7 GPa. It is important to mention that from the $\rho_{ab}(T = 260 \text{ K}, P)$ curves for PrNi₂B₂C (see inset of Fig. 6) and PrCo₂B₂C (not shown), we discarded some structural phase changes that could be related to these effects. The fact that the Pr-Ni system requires more pressure to behave almost in the same way as the Pr-Co system at low temperature could be related to changes in the *c* parameter. The main difference in the unit cell of these two compounds arises from this parameter, which is biggest in the Pr-Co system and related to the modifiable tetrahedral B-T-B angle. On the other hand, it is known that for the spin fluctuator YMn₂, the existence of a magnetic moment on Mn sites depends largely on the interatomic Mn-Mn distance [43]. Above a critical distance there exists a magnetic moment. Such distance plays a key role in determining the magnetic properties and is sensitive to external or internal perturbations. The case for Pt in PrPt₂B₂C could be similar. As we already pointed out, this compound shows an upward curvature in $\rho(T)$ at high temperature, which has been related to CEF effects [26]. We made resistivity measurements for polycrystalline PrPt₂B₂C under several pressures up to 21.5 GPa (see Fig. 7). As can be observed, the negative curvature of $\rho(T)$ at high temperature is not appreciably modified under pressure and the main changes are at low temperatures. The superconducting transition temperature, $T_{\rm c}$, was taken as the value of the 50% resistivity drop. Here, it is important to mention that the transition width for all the curves is about of 0.8 GPa, except for the second curve corresponding to 3.0 GPa, where it is about 1.0 GPa. This effect can be due to the polycrystalline nature of the sample, but we do not discard some internal stresses. According to



Figure 6 (online color at: www.pss-a.com) Normalized R(T)/R(40 K) curves at several pressures up to 2.5 GPa for polycrystalline PrNi₂B₂C. Inset shows the behavior of $\rho(P)$ at 260 K.





Figure 7 (online color at: www.pss-a.com) R(T) curves for polycrystalline PrPt₂B₂C, measured up to 21.5 GPa. High pressure tends to destroy superconductivity. Inset shows the low-temperature behavior of R(T).



Figure 8 (online color at: www.pss-a.com) Decreasing of the superconducting transition temperature for $PrPt_2B_2C$, as a function of pressure. The rate of decreasing of the transition looks normal for a d electronic compound.

Fig. 8, T_c decreases at the rate $dT_c/dP = -0.34$ K/GPa. It seems that there is no correlation between the decreasing of T_c and the unmodified curvature related to CEF effects. A positive magnetoresistance at low temperature in this compound has been associated with spin fluctuations [26]; however, although this picture follows the same trends of Pr(Ni,Co)₂B₂C, further investigations are required in order to clarify this matter.

4 Conclusions High pressure resistivity measurements in $Pr(Co,Ni,Pt)_2B_2C$ have been made. The first and foremost fact we found is that applied pressures of about 4.0 GPa are able to change drastically the low temperature resistivity behavior of $PrCo_2B_2C$, but it requires ~13.0 GPa

in order to attain similar changes for PrNi₂B₂C. This means that the low temperature electronic transport properties of PrCo₂B₂C are more pressure sensitive than those of the isomorphous PrNi₂B₂C. Evidence for spin fluctuation in PrCo₂B₂C is reported from the quadratic behavior of its resistivity at low temperature, and from the decreasing of the quadratic coefficient as a function of pressure. For PrCo₂B₂C, the magnetic scattering related to spin disorder is suppressed at 2.9 GPa, but it remained observable at less than 5.3 GPa for PrNi₂B₂C. For the case of the superconductor PrPt₂B₂C, pressure does not modify the $\rho(T)$ curvature related to CEF effects, but decreases $T_{\rm c}$ at the rate $dT_c/dP = -0.34 \text{ K/GPa}$. Finally, although these conclusions are not decisive, we believe that they would stimulate further experimental and theoretical studies for a better understanding of the pressure effects in the RT₂B₂C compounds, which is far for complete.

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