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Pressure effects in $\Pr T_2B_2C$ (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 2K. At room pressure the $\rho_{ab}(T)$ curves for the non superconducting 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 fluctuations mechanism. In the other hand 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.

PACS numbers:

I. 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 intermetallics compounds, with different rare earths (R) and transition metal (T) combinations, have been synthesized [5, 7–14] and most of them, such as HoNi₂B₂C [15], show superconductivity in spite of the presence of the rare earth magnetic element.

The influence of the transition metal magnetism in the magnetic properties of these compounds, seem 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 [16], and electronic transport measurements [11, 17], have revealed that no significant magnetic moment develops in the T sites. Local structure studies at the Ni site, using Mossbauer spectroscopy on 57 Fe doped (1 at %) samples, also support this fact [18]. Interestingly, the T elements play an indirect role in the magnetism of magnetic RT_2B_2C systems through the spatial dependent indirect RKKY exchange interac-

tions [6, 19], that govern the magnetic ordering in these compounds. On the other hand, the electronic influence of the T elements in the superconductivity of RT_2B_2C 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 [20]. On the contrary, in a comparative study of the structure and superconducting properties of RNi_2B_2C , Loureiro *et al.* [20], showed that the superconducting state is more strongly affected by the magnetism of the R ion than for the Rion 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 RT_2B_2C when magnetic R ions are presents, is not clear yet. In this work we aboard the study of a particularly interesting case: the PrT_2B_2C 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 [21], however, PrPt₂B₂C does superconduct at 6 K, even in the magnetic Pr^{+3} ion presence [22, 23]. Noticeably, $PrPt_2B_2C$ does not show any magnetic ordering at low temperatures [23] but in contrast, Pr-Ni and Pr-Co based borocarbides, develop antiferromagnetic ordering at about 4 and 8.5 K, respectively [24, 25]. Recently, magnetoresistance and specific heat studies in $Pr(Co, Pt)_2B_2C$ [25, 26] have pointed out that 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 could be not so clear. High pressure experiments in spin fluctuators such as RCo_2 [27], $CeNi_5$ [28], and UPt₃ [29] 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: $Pr(Ni, Co, Pt)_2B_2C$. We analyzed the changes of the resistivity as a function of pressure and temperature. We assume that interactions between itinerant electrons plays an important role in the low temperature resistivity characteristic, and those can be modified by applied external or internal chemical pressure.

II. EXPERIMENTAL DETAILS

Three compounds were prepared: samples of PrCo₂B₂C, PrNi₂B₂C, and PrPt₂B₂C. The single crystals were grown by cold copper crucible method as described by Durán et al [25]. All samples were characterized by X-ray diffraction using a Bruker P4 diffractometer, with monochromatized Mo-K α radiation. The cell parameters were: a = 3.6156 Å, and c = 10.3507Å for $PrCo_2B_2C$, a = 3.6996 Å, and c = 9.9885 Å for $PrNi_2B_2C$, and a = 3.8373 Å, c = 10.761 Å for $PrPt_2B_2C$ 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 (cell 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 $80 \times 15 \times 40 \ \mu m^3$ and were placed in the gasket hole. The transmitting pressure medium was MgO powder. The metallic gasket was electrically insulated, pressing over it Al₂O₃ powder of 1 μm 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 aluminum foil placed under the four gold wires, with this set up frequently we reached high quasi-hydrostatic pressures in the range 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 $Pr(Ni, Pt)_2B_2C$ 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 were used.

FIG. 1: (Color online) 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.

III. RESULTS AND DISCUSSION

It is known that the RNi₂B₂C compounds crystallize in the tetragonal body-centered structure (space group I_{4} /mmm) and when the rare earth atom radii is increased (R goes from La to Lu), the c-parameter becomes larger whereas the *a*-parameter decreases [16, 30]. This structural behavior can be accounted by for the rigidity of the B-C and Ni-B bonds and the variable tetrahedral angle in the NiB_4 unit. Distortions of this tetrahedral unit are claimed to be a decisive parameter for Tc in non magnetic or antiferromagnetic RNi₂B₂C and RNiBC compounds [31]. 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. Fig. 1 displays the T-T shortest length between 3d ions, particularly in the Pr(Ni, Co, Pt)₂B₂C 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 radii T-size causes an increasing of the *c*-parameter and an anomaly behavior of the *a*-parameter for the PrCo₂B₂C 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 radii size of the T element.

Fig. 2 shows the normalized electrical resistivity in

FIG. 2: (Color online) Normalized resistivity at 295 K $\rho_{ab}(T)/\rho_{ab}(295 \text{ K})$) of PrNi ₂B₂C, PrCo₂B₂C, and PrPt₂B₂C compounds at room pressure. The three systems present metallic conductivity from room temperature to down 25 K. Inset shows the low temperature variation of the normalized resistivity.

the a-b plane as a function of temperature for $PrNi_2B_2C$, PrCo₂B₂C, and PrPt₂B₂C single crystals at room pressure. The three compounds present metallic characteristic. At low temperature 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 ratios RRR for the first two compounds are 9 and 33 respectively, whereas for $PrPt_2B_2C$ is 5.5. The residual resistivity ρ_0 of all three is sample dependent, varying between 5 and 25 $\mu\Omega cm$. 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 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 comes to disturb 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 RNi_2B_2C series, and it has been associated with a decrease of the magnetic scattering of the conduction electrons by rare earth ions ([32] and reference therein). However, according to the results present below, it is possible that other 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 6 K. Magnetic and heat capacity measurements in this compound [23], have revealed a nonmagnetic ground state for Pr ions due to CEF effects, which is claimed to be the reason for superconductivity. The inset of the Fig. 2 shows the resistivity behavior from

FIG. 3: (Color online) Linear fit to $\rho(T^2)$ from 2 to 8 K for PrCo₂B₂C. The low temperature behavior of $\rho(T)$ follows a T^2 low with a cuadratic coefficient A = 0.08 $\mu\Omega cm/K^2$. Inset shows the $\rho(T)$ behavior at low temperatures.

60 to 2 K for the three compounds at room pressure. At first glance, increasing the transition metal radius corresponds to a major resistivity droop 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 cuadratic coefficient A, equal to 0.08 $\mu\Omega cm/K^2$, see Fig. 3. This low-temperature resistivity behavior is similar to the observed in heavy fermion systems, as for example; $YbNi_2B_2C$ and UPt_3 compounds [33, 34], and could be attributed to spin fluctuations [26, 35]. Thus, in a similar compound but simpler, RCo_2 , a T^2 dependence has been found at low temperatures, which is due to spin fluctuating characteristics [27]. The fact that the magnitude of the cuadratic coefficient A of $\rho(T)$ for PrCo₂B₂C is of the order of that for RCo_2 ([36], found this coefficient as big as three orders of magnitude but in polycrystalline $PrCo_2B_2C$) suggests that spin fluctuating could be the responsible mechanism for the low temperature $\rho(T)$ behavior in this compound. Using the universal relation for heavy fermion compounds; $A/\gamma^2 = 1.0x10^{-5}\mu\Omega$ $cm(mole K)^2/mJ^2$ [37] the resulted Sommerfeld coefficient is $\gamma = 89.4 \ mJ/mol - K^2$ which is a low value compared with that for $PrNi_2B_2C$ (211mJ/mol-K² [25] by specific heat measurements), but enhanced value as compared to the normal metal Co and to other borocarbides as YCo₂B₂C [11], (Gd, Tb, Dy, Ho, Er, Tm)Ni₂B₂C [38], whose γ is about 17 $mJ/mol-K^2$.

Figs. 4 and 5 shown 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 $\rho_{ab}(T)$ FIG. 4: (Color online) The graph shows pressure effects on the $\rho_{ab}(T)$ for a PrNi₂B₂C single crystal up to 5.3 GPa. Vertical line indicates the increasing pressure. Insert is a zoom of $\rho_{ab}(T, P)$ at low temperature.

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 room temperature) is attributed to electron-phonon scattering and under the applied pressures, it shows a slope decreasing from 0.089 $\mu\Omega$ cm/K 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 the decrease of magnetic scattering [32], 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 more stronger than in PrNi₂B₂C, mainly in the low temperature regime. From room pressure to about 1.7 GPa the high temperature behavior of $\rho_{ab}(T)$, from 300 K to about 75 K, remains without appreciable changes and with almost a constant slope of 0.30 $\mu\Omega cm/K$. In an opposite way to $PrNi_2B_2C$, 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. [36] applying pressures up to about 1.2 GPa, the maximum pressure value they applied. Similar to their results, we also observed that the cuadratic behavior of $\rho_{ab}(T)$ at low temperatures is maintained under 1.2 GPa. However, we found, applying pressures higher than 1.7 GPa on 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** in Fig.5). Interestingly, at this pressure the T^2 behavior disappeared and instead there is an appearing of a type plateau zone in $\rho_{ab}(T)$ which start about 15 K and extend down to 1.8

FIG. 5: (Color online) In a) it is presented $\rho_{ab}(T)$ for $PrCo_2B_2C$ 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.

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 K 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 $\mu\Omega \mathrm{cm/K}$.

The above experimental facts reveal that the electronic properties of $PrCo_2B_2C$ are more pressure sensitive than that for $PrNi_2B_2C$, mainly at the low temperature regime. Interestingly, we note that the shape of $\rho_{ab}(T)$ for $PrCo_2B_2C$ under pressure tends to be qualitatively similar to that for nonmagnetic YCo_2B_2C at room pressure [11]. Thus, it seems the effect of pressure in this compound is to suppress the magnetic correlations which originate the low temperature scattering behavior.

According to Fig 5, the resistivity can be fitted to a T^2 behavior only for a region of relatively low temperatures, interestingly under pressure the fitting range is extended. As already we pointed out, at atmospheric pressure the fitting goes from 2 up to about 8 K with a cuadratic coefficient A, equal to 0.08 $\mu\Omega \text{cm/K}^2$. 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 not more possible to fit a cuadratic function to the curvature of $\rho_{ab}(T)$ because the like plateau 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 by the spin FIG. 6: (Color online) Normalized R(T)/R(40 K) curves at several pressures up to 2.5 GPa for polycrystalline $PrNi_2B_2C$. Insert shows the behavior of $\rho(P)$ at 260 K.

fluctuation scenario [39], which also take in account the decreasing of the A parameter with the applied pressure. Additionally, relatively low magnetic fields decreases the A parameter in a linear form in $PrCo_2B_2C$ [26], which has been claimed to be due to quenching of the spin fluctuation. Similar results have been found in other systems as for example, Ce_{0.8}(Pr, Nd)Ni₅, indicating that spin fluctuations tends to be suppressed both by pressure and magnetic fields [40, 41]. 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 last tends to be a real non-magnetic state; one where no microscopic magnetic moments exists. This is important because in the spin fluctuations model the state above T_{sf} (where the spin fluctuation appears and which coincide with the temperature below which a T^2 law in resistivity is valid) is non the non magnetic state (like in the stoner model) but a magnetic state, where local moments still exist, but long range order tend 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 they can be suppressed by two factors: intense magnetic field and/or external applied pressure. Pressure increases the correlation there exist between f ions and promotes the itinerance of f electrons. As a result the fdensity 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 the $\rho(T)$ of PrNi₂B₂C could follow a similar behavior to that for its isomorphs PrCo₂B₂C at higher pressures, we carried out measurements for polycrystalline sample at several pressures up to 21.5

FIG. 7: (Color online) R(T) curves for polycrystalline PrPt₂B₂C, measured up to 21.5 GPa. High pressure tends to destruct superconductivity. Insert shows the low temperature behavior of R(T).

GPa. Interestingly, we found there is a marked tendency of $\rho(T)$ at low temperature to behave similar to that for $PrCo_2B_2C$ (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 Pr-Co system but at about 1.7 GPa. It is important to mention that from the $\rho_{ab}(T = 260K, P)$ curves for PrNi₂B₂C (see inset of Fig. 6), and $PrCo_2B_2C$ (not shown), we discarded some structural phase changes that could be related to these effects. The fact that Pr-Ni system requires more pressure to behaves almost in the same way to 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 stands from this parameter, which is biggest in 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 [42]. 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 point out this compound shows an upward curvature in $\rho(T)$ at high temperature which has been related to crystalline electric field (CEF) effects [25]. We make resistivity measurements for polycrystalline PrPt₂B₂C under several pressures up to 21.5GPa (see fig. 7). As it 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, Tc, decreases at the rate dTc/dP= -0.34 K/ GPa (see Fig. 8). It seems there is not correlation with the decreasing of Tc and the unmodified

FIG. 8: (Color online) Decreasing of the superconducting transition temperature for $PrPt_2B_2C$, as function of pressure. The rate of decreasing of the transition looks normal for a *d* electronic compound.

curvature related to CEF effects. A positive magnetoresistance at low temperature in this compound has been associated with spin fluctuation [25], however although this picture follows the same trends of $Pr(Ni,Co)_2B_2C$, further investigations are inquired in order to clarify this matter.

IV. CONCLUSIONS

High pressure resistivity measurements in $Pr(Co,Ni,Pt)_2B_2C$ has been made. The first and

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foremost fact we found is that applied pressures of about 4.0 GPa are able to change drastically the low temperature resistivity behavior of PrCo₂B₂C, 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 that for the isomorphous PrNi₂B₂C. Evidence for spin fluctuation in PrCo₂B₂C is reported from the cuadratic behavior of its resistivity at low temperature, and from the decreasing of the cuadratic coefficient as a function of pressure. For $PrCo_2B_2C$, the magnetic scattering related to spin disorder is suppressed at 2.9 GPa, but it remained observable at less under 5.3 GPa for $PrNi_2B_2C$. For the case of the superconductor PrPt₂B₂C, pressure does not modify the $\rho(T)$ curvature related to CEF effects, but decreases Tc at the rate dTc/dP = -0.34 K/GPa. Finally, although these conclusions are no decisive, we believe they would stimulate further experimental and theoretical studies for a better understanding of the pressure effects in the RT_2B_2C compounds, which is far for complete.

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