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Spin Fluctuations and Itinerant Magnetism in $PrCo_2B_2C$ Compound

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Abstract Resistivity as a function of the temperature and applied magnetic field was studied in $PrCo_2B_2C$ intermetallic compound. The resistivity as a function of temperature at different magnetic fields shows a parabolic behavior at low temperatures which persists for magnetic fields up to 7.8 T. Magnetoresistance measurements show negative values at temperatures below 15 K and low fields, and becomes positive at high fields. Specific heat measurements reveal a high value of the Sommerfeld coefficient of about 300 $\mathrm{mJ(mol\,K^2)^{-1}}$. These results at low temperatures are interpreted from the point of view of the spin fluctuations theory.

Keywords Spin fluctuations · Borocarbides · Magnetism · Superconductivity

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1 Introduction

The compounds with formula RNi_2B_2C (R is a rare earth or Y) have attracted much attention because they show many interesting physical properties, the most important being superconductivity and/or magnetism. The superconducting compounds have been studied intensively and much experimental and theoretical work has been reported [1]. One characteristic of these compounds is that the rare earths and not Ni

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are the responsibles of the magnetic behavior through an indirect exchange coupling of the Ruderman-Kittel-Kasuya-Yosida (RKKY) type [2, 3].

When Ni is replaced by another $3d$-element in this family of compounds, for instance cobalt atoms, they form again a solid solution with isomorphous structural characteristics to the compound with Ni. The partial substitution of Ni by Co in the superconducting borocarbides suppresses superconductivity. It is argued that this suppression is due to a decrease of the density of states at the Fermi level and a weakening of the electron-phonon coupling [4, 5]. However, the richness of the magnetic behavior persists in the RCo_2B_2C compounds. From the structural point of view, Co substitution produces changes in the tetragonal unit cell parameters respect to the nickel based borocarbides. The c axis stretches whereas the a axis shortens, and the cell volume decreases. Also the Co–Co distance gets smaller, and the separation between the magnetic R planes increases [6]. Experimental studies show that the magnetic transition temperature in each RCo_2B_2C compound is lower than in the corresponding RNi_2B_2C compound. Also, the de-Gennes factor does not scale with the magnetic transition temperature. In particular, $PrCo_2B_2C$ does not follow the general trend of the other compounds of the same family. For example, the magnetic transition is exceptionally broad and its $T_N = 10$ K is higher than the transition temperature observed in $PrNi_2B_2C$, $T_N \approx 4$ K.

Rare earth and $3d$ element compounds form magnetic systems with unusual behavior [7, 8]; for example, the RCo_2 compound shows an enhancement of the Pauli susceptibility χ_0, as well as the Sommerfeld coefficient γ. It is believed that such behavior is due to spin fluctuations (SF), which are produced by the Co $3d$ electrons [7, 8]. It is important to mention that one of the characteristic feature of systems with spin fluctuation is obtained by observing the behavior of the resistivity vs temperature curves, $\rho(T)$. This is characterized by a T^2 dependence at low temperatures, and saturation at high temperatures. Due to the magnetic character of these systems, when a magnetic field is applied, the magnetoresistance (MR) will be negative and proportional to the square of the applied magnetic field [9–11].

Examining the behavior of the family of RCo_2B_2C compounds, El Massalami et al. [6, 12] have reported that $\rho(T)$ in the RCo_2B_2C family shows a typical metallic behavior at high temperatures and an incoherent scattering contribution associated with the crystal-electric field (CEF) and hybridization influences at low temperature. Studies on single crystals of (Y, Lu)Ni_2B_2C show that the temperature dependence of the transverse MR presents a T^p behavior where $p = 2.2$ and 2.0 for Y and Lu compounds, respectively. In spite of this behavior, it is not very clear if this compound may be catalogued as a spin fluctuating system. There was suggested that the T^2 characteristic could be produced by electron-electron scattering and by another type of scattering mechanisms [13]. A different situation in the in-plane longitudinal MR of $HoNi_2B_2C$ was found, where the negative MR was associated to the spin-disorder scattering [14]. Moreover in other compounds of this family, for example the $Ce(Ni_{1-x}Co_x)_2B_2C$, the MR is positive at 4.2 K at low Co content, but between 0.75 to 1 Co content and below 1 T the MR is negative, changing to positive at higher fields. In this case the negative MR has been associated to an antiferromagnetic (AF) impurity, like CeB_2C_2, while the positive MR is the result of electron scattering by the applied field [15]. Accordingly, the electrical resistance characteristics of these

compounds are still not very clear, however, these results suggest that spin fluctuations is a process that can be present in some Co based borocarbides.

In this work, we report studies on the temperature and magnetic field dependence of resistivity in the $PrCo_2B_2C$ compound, at low temperatures. It was found that the resistivity as a function of temperature with and without magnetic field shows a quadratic behavior for temperatures below 5 K. This behavior is analyzed from the point of view of the spin fluctuation scenario. In addition, heat capacity measurements show that the Sommerfeld coefficient γ has an enhanced value just before the magnetic transition.

2 Experimental Details

Pure samples of $PrCo_2B_2C$ were obtained and characterized as reported by Durán et al. [16]. In short, the samples were prepared in a radio-frequency induction furnace, melting a mixture of the appropriate quantities of Pr (99.9%), Co (99.9%), B (99.8%) and C (99.9998%), in an Ar flow. From a powder x-ray diffraction pattern the cell parameters, $a = 3.61560(1)$ Å and $c = 10.3507(6)$ Å were determined, coinciding with the reported values [6]. The polycrystalline sample studied has dimension of about $2.5 \times 1.5 \times 1.5$ mm^3. The ac resistance measurements were carried out using four 25 μm diameter gold wires attached with silver paste to the specimen, the injected RMS current was about 10 mA. Magnetoresistance data were taken at constant temperature and at magnetic fields from 0 to 7.8 Tesla, in perpendicular direction to the applied current. Above 4.2 K, the temperature was stabilized using a regulating system with a capacitance thermometer as sensor. Below 4.2 K, the vapor pressure of the helium bath was controlled and used as thermometer. Magnetization measurements were taken in a SQUID based magnetometer MPMS-5S (Quantum Design). The specific heat (C) measurements were performed in a PPMS (Quantum Design) calorimeter using the two-τ relaxation method. The sample was glued on the platform using Apiezon N grease. Once measured the total heat capacity consisting of sample, platform, and grease was determined. The sample heat capacity was determined subtracting the platform and grease contributions.

3 Results and Discussions

Resistivity $\rho(T)$ without magnetic field, and magnetization $M(T)$ of $PrCo_2B_2C$ are shown in Fig. 1. From 100 K to about 7 K, $\rho(T)$ shows a linear behavior that disappear between 6 and 7 K. At lower temperatures the main feature observed in $\rho(T)$ shows a quadratic shape as is displayed in Fig. 1a. The continuous line is a fit to the experimental data between 1.2 K and 5 K, the best fit was obtained using $\rho(T) = \rho_0 + AT^2$ with $\rho_0 = 61$ μΩ cm and $A = 0.053$ μΩ cm K^{-2}. The $\rho(T)$ general trend shows a metallic-like behavior as is observed in the inset of Fig. 1a. The quadratic fit is quite good, near to the temperature where a maximum in the magnetization measurements is observed. However it is below 10 K, where the antiferromagnetic transition temperature was reported for this compound [6].

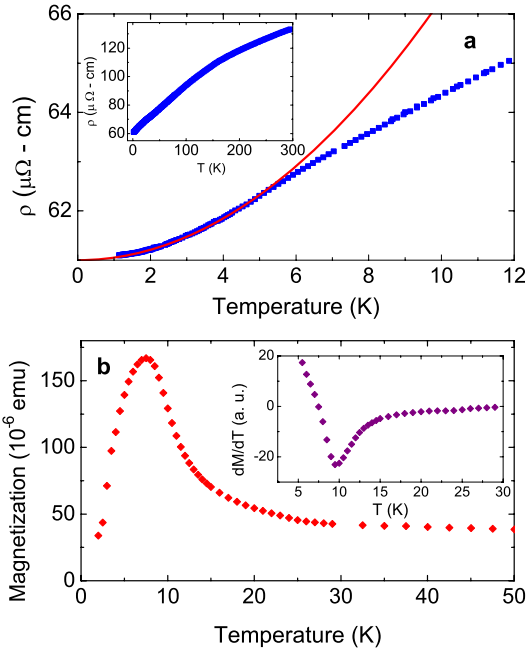

Fig. 1 (Color on-line) (**a**) $PrCo_2B_2C$ resistivity as a function of temperature showing a quadratic dependence (*continuous line*) at low temperature. *Inset* shows $\rho(T)$ between 1.2 K and room temperature. (**b**) Magnetization as a function of temperature at low temperatures, the maximum is displayed at about 8 K. The *inset* shows the temperature derivative of the magnetization. Note the minimum at 10 K associated with the antiferromagnetic transition temperature

Measurements of the magnetization as a function of temperature were performed in zero field cooling mode at 0.01 T (Fig. 1b). $M(T)$ shows a broad peak with the maximum at about 8 K, in agreement to reported measurements [17]. This magnetic anomaly was characterized by ac susceptibility measurements as an antiferromagnetic transition with the maximum at 10 K [6]; however, this broad peak is not as the sharp peak at the onset of the AF ordering observed in the Ni-borocarbide compounds [18], in fact, as far as we know the magnetic order has not been determined unambiguously. The inset in Fig. 1b shows the derivative of $M(T)$ as a function of temperature, there the minimum at 10 K is in coincidence with the antiferromagnetic transition temperature observed by magnetization and heat capacity measurements [6, 12, 19].

Specific heat (C) measurements at low temperature, presented as C/T vs. T^2 in Fig. 2, do not show the characteristic λ type magnetic transition, however, a tiny feature was observed near 10 K which is in agreement with the $M(T)$ and specific heat measurements reported in the literature [6, 12]. Comparing our measurements with the results reported in [6, 12], there are several differences. The magnetic transition is not well defined in both measurements, however in the work reported by El Massalami et al. there is a smeared shoulder around 10 K that gives more evidence of the magnetic transition. Another difference is that the heat capacity values in our measurements are lower than their reported values. We believe that these differences, particularly the higher specific heat values, may be due to an extra contribution to the heat capacity, which may be produced by a magnetic impurity or a minor phase. It is noted that C_M above 10 K [12] is almost constant instead to decrease, implicating the possibility of another magnetic contribution.

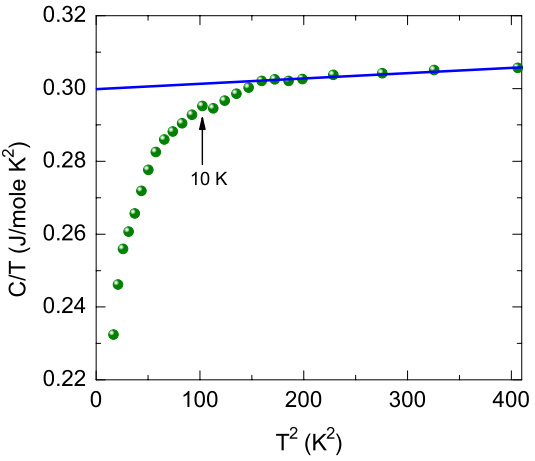

Fig. 2 (Color on-line) Heat capacity of $PrCo_2B_2C$ at low temperatures. The *line* is a least square fitting of the data between 14 K and 20 K

Assuming a simple approach, $C = \gamma T + \beta T^3$, we performed a least squares fitting of the experimental data between 14 K and 20 K. This temperature range was taken in order to avoid the magnetic specific heat tail above the magnetic transition. From such fitting a Sommerfeld coefficient, $\gamma = 299$ mJ(mol K^2)$^{-1}$ was obtained, however, this value must be considered a crude approximation. It is worth emphasizing that the value obtained for the γ coefficient in $PrCo_2B_2C$ implies a heavy fermion compound, and of the same order than $\gamma \approx 250$ mJ(mol K^2)$^{-1}$ reported for the isomorph $PrNi_2B_2C$ compound [1, 20]. It is important to mention that as result of the fitting, the value of Θ_D is 929 K, which is higher than the values reported for other borocarbides [4, 21–24]. There Θ_D ranged from 300 K to 500 K. Moreover, in the $Y(Ni_{2-x}Co_x)B_2C$ compound has been observed that Θ_D increases as the content of Co is increased [4]; from 480 K for $x = 0$ to 500 K for $x = 0.4$. On the base of this tendency, it is expected that Θ_D in $PrCo_2B_2C$ must be higher than the Θ_D values of other borocarbides. We do not discard that above the magnetic transition temperature a magnetic contribution is present, due to the tail of the magnetic transition. We think that this contribution decreases rapidly with temperature and then its effects can be neglected.

Some particular characteristic of $PrCo_2B_2C$ and $PrNi_2B_2C$ may help to better understand the behavior of this Co based compound. For instances, it has been reported that the AF transition in $PrNi_2B_2C$ occurs at about 4 K, whereas in $PrCo_2B_2C$ occurs near 10 K. In this respect, it is accepted that the RKKY interaction, the crystalline electric field effects, as well as the Pr -f and the Ni/Co -d electronic bands hybridization, are responsible of the magnetic behavior and the anomalous features observed at low temperatures [12, 16, 25]. Thus, the difference of the AF transition temperature in $PrCo_2B_2C$ and $PrNi_2B_2C$ compounds may be produced by changes in the interatomic distances [6, 17]. Thus, Co substituting Ni, results in a modifications of the RKKY interaction and the crystalline electric field effects. In addition, recent band structure calculations [26] in $PrCo_2B_2C$ compound showed an appreciable hybridization between the admixture of Pr -f, Co -d and B -p states at the Fermi energy level. It is clear that this appreciable overlapping is the responsible of the

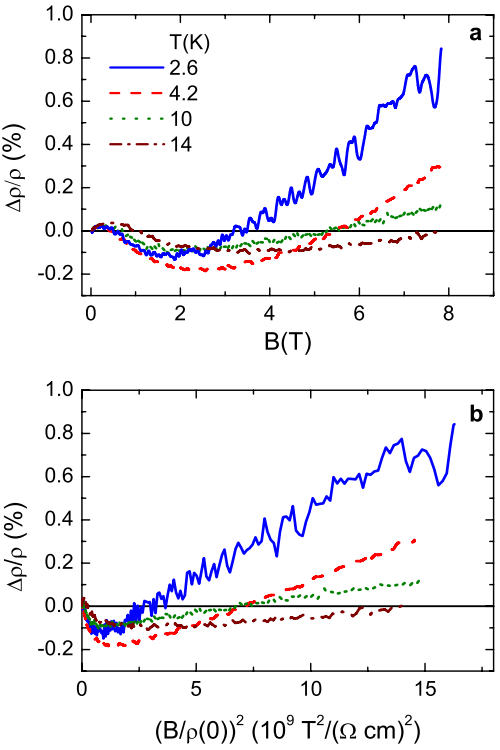

Fig. 3 (Color on-line) (**a**) Normalized magnetoresistance as a function of applied magnetic field at various temperatures. (**b**) Magnetoresistance against $(B/\rho(0))^2$ where the linear trend above 3 T is in agreement with the Kohler's rule

electronic enhancement of the specific heat observed just before the magnetic transition temperature, which can give rise to an extra contribution of f electrons at the Fermi level. This fact, then increases the Sommerfeld constant giving rise to a heavy fermion system.

Figure 3a presents the normalized magnetoresistance $\Delta\rho(B,T)/\rho(0,T) = [\rho(B,T) - \rho(0,T)]/\rho(0,T)$ for $PrCo_2B_2C$ as a function of the applied magnetic field, at various temperatures. The magnitude of the MR shows a small positive value between 0 and 0.5 T. As the field is increased MR goes to negative values and, at higher fields, it is positive again. For instance, the MR curve measured at 2.6 K has positive values for fields above 3 T. Magnetoresistance versus $(B/\rho(0))^2$ is presented in Fig. 3b, in this representation MR follows a linear trend above 3 T, that means that MR has a quadratic behavior with magnetic field. This behavior is in agreement with the Kohler's rule, however, in these curves it is possible to consider two contributions; a positive contribution due to the cyclotron motion of the conduction electrons and a negative contribution that could be due to a magnetic order or spin fluctuations [11].

In order to analyze the $\rho(T)$ behavior at temperatures below 5 K, in Fig. 4 we plotted the resistivity as a function of T^2, at different applied magnetic fields. The continuous lines are a least square fitting of the data, so that the resistivity is described by the expression,

$$\rho(B,T) = \rho_0(B) + A(B)T^2.$$

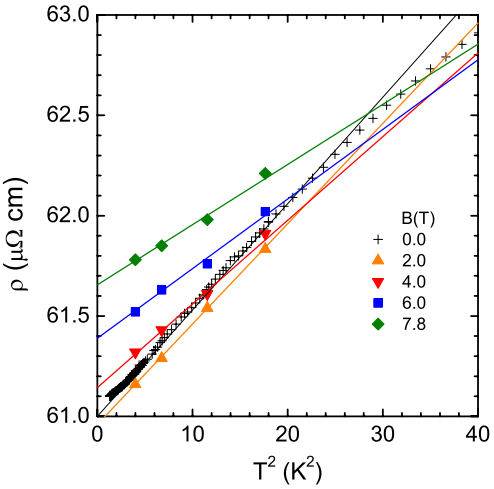

Fig. 4 (Color on-line) Resistivity as a function of T^2 at different applied magnetic fields. *Continuous lines* are a least square fit of the data

The first term is the residual resistivity, due to impurities and lattice faults (in a magnetic or nearly magnetic metal this term includes a spin fluctuation contribution). The second term can be related to electron-electron and/or electron-SF scattering [11, 27]. According to the Mathiessen's rule, a term related to phonon scattering must be included, but in this case it was not considered because of this contribution is negligible for temperatures below 5 K.

From the least-square fitting data ρ_0 and A values were obtained. These coefficients are plotted in Fig. 5, as a function of the applied field. We observe a minimum in ρ_0 at about 1.7 T and, from about 2 T ρ_0 increases almost linearly up to 7.8 T. The variation of $\rho_0(B)$ is small, only about 1 percent on the range of fields investigated. On the other hand, A decreases by a factor of 2 in almost a linear form, for field about 1 T up to 7.8 T. The decreasing of $\rho_0(B)$ at low field, and the linear behavior of $A(B)$ are characteristic features of weakly ferromagnetic metals, where the temperature quadratic behavior of resistivity is originated by spin fluctuations [9]. Moreover, the decreasing of $A(B)$ indicates that spin fluctuations are quenched by the magnetic field [9, 11]. This similar behavior has been observed in ρ_0(B) and A(B) in single crystals of the heavy fermion borocarbide, $YbNi_2B_2C$ [28]. The residual resistivity, measured with the magnetic field applied perpendicular to the current and to the c axes, increases and the coefficient of the quadratic term decreases as the field is increased. In that compound the parabolic temperature behavior of the resistivity was observed below $\sim$1.6 K. The MR below this temperature is anisotropic, being positive when the field is applied perpendicularly to the c axes. It shows a maximum at about 10 T and changes to negative values as the field is increased. When the field is parallel to the c axes, MR is negative and shows a relative minimum. The MR behavior in this compound was explained as a result of the electronic scattering by charge fluctuations, produced by ligand disorder [28]. The disorder is produced when the intermetallic ion is replaced by impurities. A second kind of disorder was considered when substitutional impurities on the f-electronic sites are present. It is called a Kondo-hole disorder. Yatskar et al. [28] claim that both type of disorder were present in $YbNi_2B_2C$, but the ligand disorder was the dominant one.

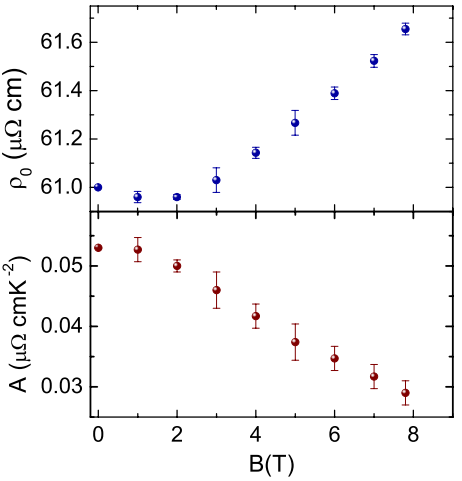

Fig. 5 (Color on-line) Magnetic dependence of the residual resistivity ρ_0 and the coefficient of the quadratic term of the resistivity A, determined from $\rho(T)$ between 2 K and 5 K

Comparing the low temperature ($T < 5$ K) behavior of $\rho(T)$, at fields below 8 T, between $YbNi_2B_2C$ and $PrCo_2B_2C$, we can get some indications about the role of disorder in $PrCo_2B_2C$. Some differences can be pointed out: (a) $YbNi_2B_2C$ has a positive MR, conversely, in $PrCo_2B_2C$ the MR is negative at low fields, changing to positive for fields above 3 T. (b) In $PrCo_2B_2C$ $\rho_0(B)$ decreases at low fields, whereas in $YbNi_2B_2C$ it increases for all applied fields. (c) $A(B)$ is almost linear in $PrCo_2B_2C$, whereas it is not linear in $YbNi_2B_2C$. These differences permit us to discard the disorder, as the main mechanism responsible of the magnetoresistive behavior of $PrCo_2B_2C$.

$PrCo_2B_2C$ presents at low temperatures a quadratic resistive behavior, that may be identified by two contributing effects: electron–electron and electron–phonon processes, those we are associating with a spin fluctuation phenomena. Also It is very well known that in transition and rare earth metal or alloys there are two or more separate scattering mechanisms contributing to the resistive behavior, operating simultaneously, and that the effect of impurities is to smear out the true temperature dependence of the resistivity. In simple metals the effect of electron-electron scattering on resistivity may be completely neglected because momentum conservation by individual collisions. But this effect is less improbable to be neglected in transition metals and/or in f-electrons systems with d conduction electrons. Particularly in this case a T-square term in the resistance needs to be considered. However this T-square term also will have a dominant influence on the scattering of the conduction electrons by spin wave, as we are assuming occurs in $PrCo_2B_2C$. Also, in addition to two of these scattering processes, it must be considered the important magnetic influence of Co (if exist) in the resistive behavior. One consideration about the electronic behavior in this magnetic Pr and/or Co based compound, and in comparison to the different non-magnetic Ni based compound (also observed in other superconducting borocarbides), is that the magnetism in this compound may adds two new scattering processes, which tend to be important; one is the crystalline electric field (CEF) effects that combined with the Pr 4-f electrons need necessarily to be considered in the total resistive features mainly at low temperatures. Other additional effect that

requires consideration, for the possible effects on the resistivity, is the itinerant or nearly itinerant $4f$ electrons. These tends to enhance the electronic mass at low temperatures, leading to a very high heat capacity, linear in temperature. This possible transition from localized to itinerant $4f$ electrons have been explained as a Kondo resonance. Under this assumption of nearly itinerating electrons, it is necessary to have present the effects of $f-f$ orbitals overlaps, which may add another scattering mechanisms to the resistive behavior. The $f-f$ overlaps, as in Pr atoms and in this $PrCo_2B_2C$ compound, are scattering processes very difficult to discard (and not quite totally understood to be considered as important effects in a spin fluctuation problem).

This physical point can be resumed; as the fact that crystal field levels may indicate that hybridizing and conduction electrons must be explicitly taken into account when $4f$-electrons are only partially de-localized. However this is a problem actually without solution. So, in order to have a complete explanation about the effects on the resistive behavior at low temperatures when two magnetic orders are interacting as in the Pr and possible Co atoms as in this compound is a difficult task and the incorporation of scattering effects must include a treatment of spin-orbit, which added to magnetic field effects yields a band structure which account for the electrons at the Fermi level. When the scattering effects of the magnetic atoms in this compound are taking into account these partly localized states interact with the surroundings through the single-ion crystal field and magnetostatic forces, and with two-ion indirect exchange. It is interesting to mention that this effect of magnetic scattering indicates that spin correlation which arises from the strong coulomb interactions between valence electrons leads to an enhanced singlet-site repulsion which can be many times larger that the coulomb attractive pseudopotential sufficient to suppress superconductivity as in this compound.

In the Moriya's spin fluctuation theory the interaction of electrons with SF, contributes with a quadratic term to the resistivity. At the same time the linear term of the electronic heat capacity is enhanced due to a raise of the effective mass. The coefficient A is enhanced by exchange interactions and is related to the spin fluctuation temperature $T_{sf} \propto A^{-1/2}$. When a magnetic field is applied, $A(B)$ decreases as the field is increased, reaching a constant value at high fields. This saturation is because the spin fluctuations are quenched by the magnetic field. In the case of weakly ferromagnetic metals, $A(B)$ is linear in the low field range; Sc_3In is an example of a weak itinerant ferromagnetic metal [9]. In this material the T^2 resistive behavior is due to electron-SF interactions. The A value in Sc_3In is about 0.05 $\mu\Omega\,\mathrm{cm\,K^{-2}}$, at zero field [11]. We have obtained a similar value for $PrCo_2B_2C$, at zero field, $A = 0.053\ \mu\Omega\,\mathrm{cm\,K^{-2}}$. Other examples of spin fluctuation systems with similar magnitude of A are; the RCo_2 (R = Sc, Y and Lu) [7, 8], and $ZrZn_2$ [9, 11]. In this sense for $PrCo_2B_2C$, electronic structure calculations [26] shown that the Pr -f electrons are not totally localized. Rather, they are hybridized with the -d and -p states and fluctuating at the Fermi level. We believe that this quasi-free admixture of -f, -d and -p electrons at the Fermi level is the responsible of the spin fluctuation behavior in this compound, and is the origin of the high Sommerfeld constant value.

Lastly, from the values of A and γ obtained for $PrCo_2B_2C$, we determined that the Kadowaki-Woods ratio, A/γ^2, is equal to $0.59 \times 10^{-6}\ \mu\Omega\,\mathrm{cm(mole\,K/mJ)^2}$. This

value is smaller than 1.0×10^{-5} $\mu\Omega\,\mathrm{cm(mole\,K/mJ)^2}$ reported for heavy fermions systems, but similar to 0.4×10^{-6} $\mu\Omega\,\mathrm{cm(mole\,K/mJ)^2}$, reported for weak ferromagnets, strong Pauli paramagnets and spin fluctuation systems [11, 29]. As a last point we therefore may assume that CEF-induced incoherent scattering processes affects the T-square resistive behavior, however our experimental observations in this compound, and experiments by other workers in the field, analyzed in different compounds with similar features, states that this behavior may be induced by spin fluctuation.

4 Conclusions

We have performed magnetization, specific heat and magnetoresistance measurements on the borocarbide $PrCo_2B_2C$ compound. A broad peak observed by means of magnetization measurements and a tiny anomaly in the specific heat, confirmed the AF transition at about 10 K. At low applied fields, $\Delta\rho$ is negative, suggesting that the magnetic fluctuations decrease the electron scattering as the magnetic field is increased. At temperatures below 5 K the resistivity as a function of the temperature at different magnetic fields, shows a quadratic behavior. Furthermore, the residual resistivity and the coefficient of the quadratic term of $\rho(T, B)$ at low magnetic field, decreases as the field increases. All these characteristics are in agreement with a spin fluctuation scenario. The origin of this behavior could be related to the moderated hybridization of the -f, -d and -p electrons. Additional evidence that support the spin fluctuation scenario is the fact that γ reaches a value close to 300 $\mathrm{mJ/mol\,K^2}$.

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