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The role of magnetic excitations in magnetoresistance and Hall effect of slightly TM-substituted $BaFe_2As_2$ compounds (TM = Mn, Cu, Ni)

J.P. Peña[a,*], M.M. Piva[b], C.B.R. Jesus[b], G.G. Lesseux[b], T.M. Garitezi[b], D. Tobia[b], P.F.S. Rosa[b,c], T. Grant[c], Z. Fisk[c], C. Adriano[b], R.R. Urbano[b], P.G. Pagliuso[b], P. Pureur[a]

[a] Instituto de Física, Universidade Federal do Rio Grande do Sul, Av. Bento Gonçalves 9500, Porto Alegre, RS, Brazil
[b] Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, Rua Sérgio Buarque de Holanda 777, Campinas, SP, Brazil
[c] Department of physics and Astronomy, University of California, 2186 Frederick Reines Hall, Irvine, CA, USA

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ABSTRACT

We report on electrical resistivity, magnetoresistance (MR) and Hall effect measurements in four non-superconducting $BaFe_{2-x}TM_xAs_2$ (TM = Mn, Cu and Ni) single crystals with small values of the chemical substitution x. The spin density wave (SDW) ordering that occurs in these systems at temperatures $T \sim$ (120–140) K, in close vicinity to a tetragonal/orthorhombic transition, produces significant modifications in their magneto-transport properties. While in the magnetically ordered phase the MR is positive and its magnitude increases with decreasing temperatures, in the paramagnetic regime the MR becomes vanishingly small. Above the spin density wave transition temperature (T_{SDW}) the Hall coefficient R_H is negative, small and weakly temperature dependent, but a remarkable change of slope occurs in the R_H versus T curves at $T = T_{SDW}$. The Hall coefficient amplitude, while remaining negative, increases steadily and significantly as the temperature is decreased below T_{SDW} and down to $T = 20$ K. The qualitative behavior of both MR and Hall coefficient is weakly dependent on the chemical substitution in the studied limit. The experiments provide strong evidence that scattering of charge carriers by magnetic excitations has to be taken into account to explain the behavior of the resistivity, magnetoresistance and Hall effect in the ordered phase of the studied compounds. Effects of multiple band conduction also must be considered for a complete interpretation of the results.

1. Introduction

Because of its peculiar electronic properties along with its similarities with the high temperature cuprate superconductors (HTCS), the Fe-pnictide superconductors have fascinated scientists since its discovery in 2008 [1]. As the cuprates, the Fe-pnictide superconductors have parent compounds showing an antiferromagnetic ground state. Also similar to the HTCS, superconductivity in the Fe-pnictides can be achieved by doping the precursor compounds with electrons or holes. Contrasting with the cuprates, superconductivity in the Fe-pnictides may also be obtained by applying pressure [2]. Further, the precursors of the iron based superconductors are metallic and the antiferromagnetism is related to stabilization of a spin density wave (SDW) state [3,4]. Nevertheless, it is currently accepted that the simultaneous presence of both localized and itinerant moments is necessary to explain magnetism, transport and other electronic properties of these compounds [5–7]. The typical $T - x$ phase diagram of Fe-pnictides shows that the parent compounds and underdoped samples exhibit a tetragonal, paramagnetic phase at high temperatures, while an orthorhombic, antiferromagnetic phase characterizes the low-temperature state. The structural and magnetic transitions are gradually suppressed upon doping the parent compound with selected impurities, and above a certain threshold of the chemical substitution, a superconducting ground state is stabilized [2].

Among several known families of iron based superconductors, the 122 family is the most studied. One of the parent compounds of this family is the $BaFe_2As_2$, which displays a structural (tetragonal to orthorhombic) transition closely followed by a SDW phase transition at $T \sim 140$ K. Furthermore, a superconducting transition can be induced by external pressure or chemical substitutions. The substitutions can be either out or inside the Fe-As planes. The first case is represented by the partial substitution of Ba atoms by K atoms (hole doping). Substitution inside the planes are achieved by partially substituting the Fe atoms by Co, Ni, Cu, Rh or Pd [2,8–12] (electron doping), or the As atoms by P atoms (isovalent substitution). All these substitutions disrupt the magnetic

* Corresponding author.
E-mail address: jullypaola@if.ufrgs.br (J.P. Peña).

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order and, depending on the impurity concentration, induce superconductivity. However as similar results are obtained by applying external pressure, it is still controversial whether chemical substitution in fact leads to an effective charge doping and is the main factor driving superconductivity in the $BaFe_2As_2$ compound [13,14]. On the other hand, although substitutions such as Mn [15] and Cr [16] also suppress the SDW phase, the superconducting state is not observed due to the strong magnetic pair-breaking mechanism induced by these impurities.

An important feature in the electronic structure of the Fe-pnictides is the presence of both electron and hole pockets on their Fermi surface [2,8]. As a consequence, it has been difficult to identify the microscopic phenomena giving place to magnetism and many doubts remain on the mechanisms governing the electric charge transport in these systems. To shed new light on the discussion of this specific subject, we have studied electrical magneto-transport properties in several non-superconducting compounds of the Ba-122 family. In particular, we have experimentally investigated the magnetoresistance (MR) and Hall effect (HE) of $BaFe_{2-x}TM_xAs_2$ (TM = Mn, Cu, Ni) single crystal samples in the very low doping limit. We also report on resistivity versus temperature measurements at zero applied magnetic field.

In general, the transversal MR ($\mu_0 H \parallel c \perp I$, where I represents the electrical current and c is the principal symmetry axis) of 122 compounds is positive and displays an unusual temperature dependence becoming negligibly small for $T > T_{SDW}$ [17,18]. On the other hand, the longitudinal MR ($\mu_0 H \parallel ab \parallel I$ where ab refers to the planar orientation) is negative, and tough small, it is still measurable in the paramagnetic region [19]. The longitudinal MR behavior in the disordered region was explained as resulting from the spin-disorder suppression mechanism produced by the external magnetic field [19]. Conversely, no explanation is found in the literature for the temperature and field dependent behavior of the transversal MR, which is the one studied here. The Hall coefficient (R_H) shows a strong temperature dependence in compounds of the 122 family [20,21]. In particular, the absolute value of R_H has a drastic increase when the temperature decreases below T_{SDW} in non-doped and slightly doped compounds. Some attempts to explain this behavior include (i) a Fermi surface reconstruction at T_{SDW} [20–22], or (ii) the effect of conduction by multiple bands. However, in undoped and slightly doped compounds as those studied here, the magnetic transition is very sharp, in some cases considered weakly first order [23], so that the first interpretation does not adequately explain the observed variations of R_H in temperatures far below T_{SDW}. The second explanation implies that, if not accompanied by another effect, strong modifications in the electronic band structure should occur all along the magnetic ordered phase. These controversies show that the mechanisms governing the magneto-transport phenomena in the 122 Fe-pnictides are still not completely understood. In this work we present evidences that carrier scattering by magnetic excitations leading to anomalous contributions both to the MR and HE should be considered as a relevant mechanism for describing the magneto-transport properties of these systems

The results here described strongly suggest that not only multiple-band conduction, but also scattering by magnetic excitations, must be taken into account for explaining both the MR and HE in the magnetically ordered ground state of the undoped and slightly doped 122 Fe-pnictides.

2. Experimental details

Single crystals of $BaFe_{2-x}TM_xAs_2$, $x = 0$ and $x = 0.020,\ 0.012$ and 0.015 were synthesized for TM = Mn, Cu and Ni, respectively. These concentrations were estimated by Energy Dispersive Spectroscopy (EDS) and wavelength-dispersive X-ray spectroscopy (WDS) analyses. We further compared the measured SDW temperatures with predictions of the phase diagrams found in literature. This last criterion is useful because of the large uncertainties related to the employed EDS and WDS techniques in the limit of low impurity concentration. The crystals were grown by using the In-flux method as reported in Ref. [10]. None of our four samples shows superconductivity and no detwinning processes were attempted on the obtained crystals.

Table 1
Lattice parameter along the c−axis and the magnetic transition temperatures for the studied samples of $BaFe_{2-x}TM_xAs_2$ (TM = Mn, Cu, Ni).

Sample	c (Å)	T_{SDW} (K)
$x = 0$	13.021 ± 0.001	135 ± 3
$Mn_{0.020 \pm 0.006}$	13.022 ± 0.003	121 ± 3
$Cu_{0.012 \pm 0.004}$	13.011 ± 0.001	123 ± 1
$Ni_{0.015 \pm 0.005}$	13.034 ± 0.002	121 ± 2

X-rays diffraction and resistivity measurements were performed for characterization. The magnetic ordering temperature T_{SDW} was estimated from the temperature derivative of the zero field electrical resistivity curves. Table 1 displays the values of T_{SDW} and the c−axis lattice parameters extracted from the experiments above mentioned.

Electric transport measurements were carried out by using a standard four probe method. The electric contacts were made with silver epoxy and Cu wires in platelet-like crystals. The contact leads for voltage measurements were attached to the same edge of the sample for measuring the longitudinal resistance, and to opposite edges for the transversal resistance. Measurements were performed upon the application of magnetic fields with magnitude between 0 and ± 9 T and orientation parallel to the c−axis. The magnetoresistance was determined from the average $\rho_{even} = (\rho_+ + \rho_-)/2$ of the longitudinal measurement and the Hall resistivity from $\rho_{odd} = (\rho_+ - \rho_-)/2$ of the transversal voltage measurement. The term $\rho_{+/-}$ refers to the vertical direction positive/negative of the magnetic field, respectively. All the magneto-transport experiments were performed with a low-frequency AC bridge of a commercial PPMS@ platform manufactured by Quantum Design, Inc.

3. Results and discussion

3.1. Resistivity

The temperature-dependent electrical resistivity curves for the studied samples are presented in Fig. 1. The magnetic transition is signaled by a typical kink in the ρ vs. T curves. The transition temperature is clearly diminished upon chemical substitution. In addition, the absolute values of the resistivity in the substituted compounds are significantly larger than that in the parent compound. Thus in the low x limit, the studied chemical substitutions seem to play the role of scattering centers increasing the relaxation rate rather than as donor/receptor atoms increasing the electron/holes density. The inset in the (a) panel of Fig. 1 shows the residual resistivities (ρ_r) of the substituted samples compared with that of the pure compound; there we include a point obtained from Ref. [24] for a sample of $BaFe_{1.98}Co_{0.02}As_2$. The data for the substituted samples are calculated for $x = 0.01$, then normalized with respect to the residual resistivity of the pure specimen. We note that the behavior of ρ_r across the series of compounds is similar to that observed in dilute alloys, where ρ_r increases when the charge of the impurity relative to that of the host also increases because of its higher cross section.

A close look at the ρ vs. T curves in Fig. 1 near the kink observed at T_{SDW} reveals that the resistivity goes through a

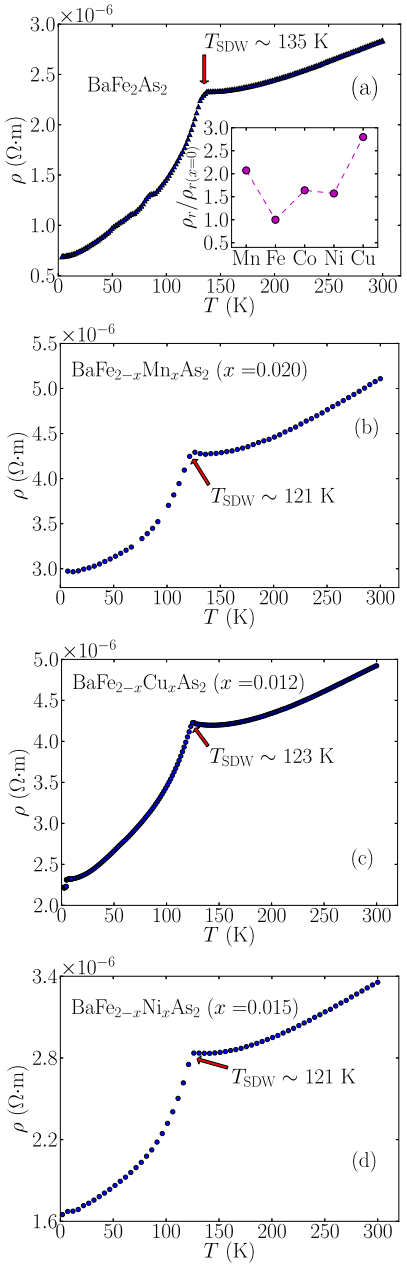

Fig. 1. Resistivity as a function of the temperature for the four studied samples. The magnetic ordering temperatures (T_{SDW}) are indicated. The inset in the (a) panel shows the ratio between the residual resistivity of the substituted-samples with respect to that of the pure compound (see text).

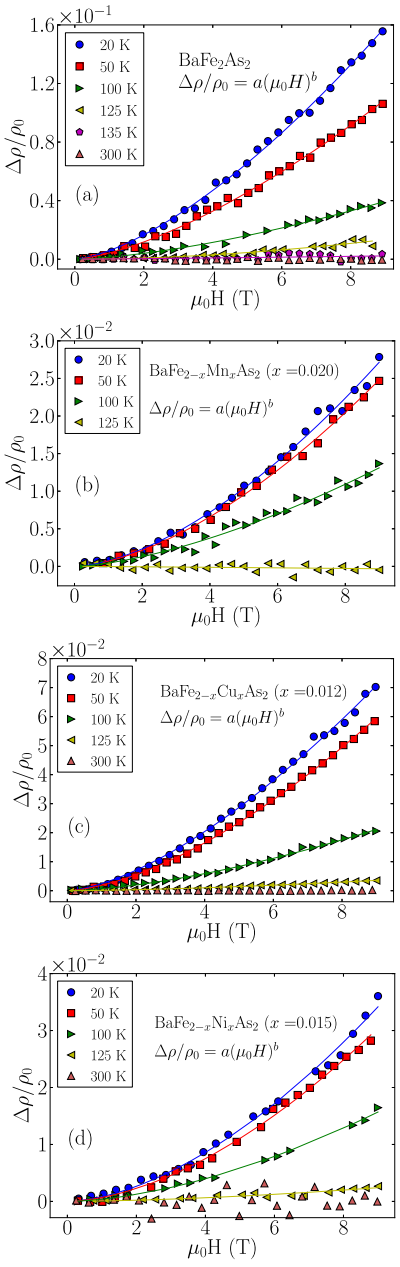

Fig. 2. MR as a function of the magnetic field (μ_0H $\parallel$ c) in several fixed temperatures. Solid lines are fits to $\Delta\rho/\rho_0 = a(\mu_0 H)^b$, where $b \approx 1.5$ for all samples and temperatures. The MR is nearly zero for temperatures above T_{SDW}.

maximum in the form of a faint cusp at this temperature. Although the magnetic transition is accompanied by a slight orthorhombic distortion in the studied compounds [2], we rather ascribe this maximum to the opening of a small gap, or pseudogap, near the Fermi level because of the antiferromagnetic ordering. This maximum is often identified as the super-zone effect, commonly observed in the resistivity of antiferromagnetic metals near the ordering temperature [25]. Alternatively, it has been argued that, in the absence of magnetic field, the kink in the ρ vs. T plot, where the resistivity drops sharply below T_{SDW}, can be explained by the existence of one or two (one for each type of carrier) successive Lifshitz transitions [26].

3.2. Magnetoresistance

In Fig. 2, the MR is shown as a function of the magnetic field at several fixed temperatures for all the studied samples. In Fig. 3 the temperature dependence of the MR is presented at the fixed fields μ_0H = 4, 6 and 8 T. In both figures the MR is given as $\Delta\rho/\rho(0)$ where $\Delta\rho = \rho(\mathrm{H}) - \rho(0)$.

Fig. 3 shows that the MR amplitude is negligible in temperatures above T_{SDW} for all studied samples. This particular result strongly suggests that the electrical transport in these systems is not a single-band conduction process. According to the simplest two-band model, the low-field MR is given as [27]:

$$\Delta\rho \approx \frac{\sigma_h \sigma_e (\mu_h - \mu_e)^2}{(\sigma_h + \sigma_e)^2} \mathrm{H}^2, \qquad (1)$$

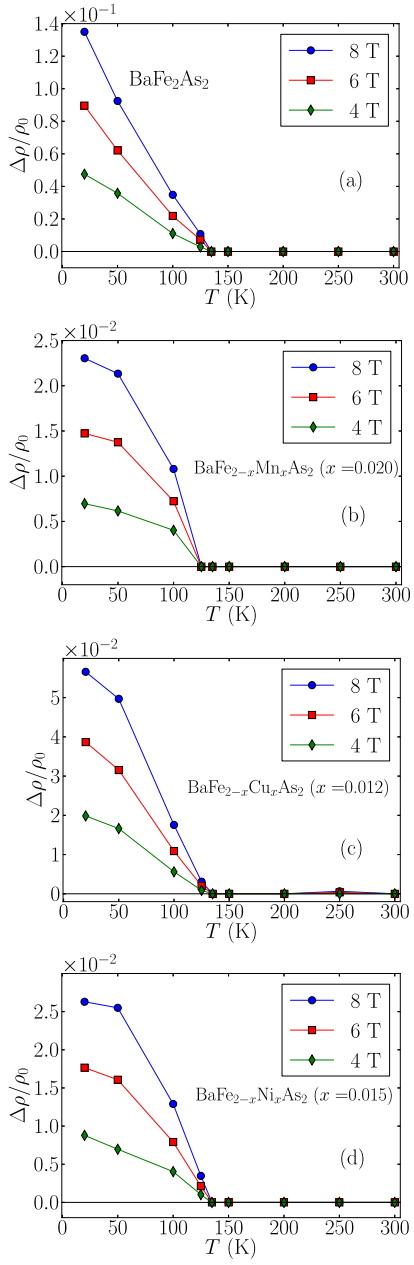

Fig. 3. MR amplitude as a function of the temperature at three different fields.

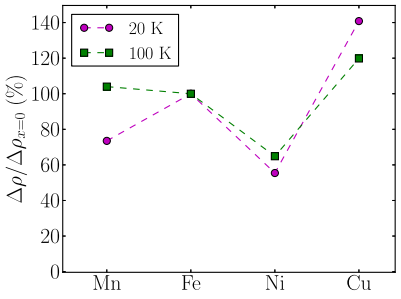

Fig. 4. Absolute magnetoresistance $\Delta\rho = \rho(\mathrm{H} - \rho(0))$ for the measured samples in $T = 20$ K and 100 K under the applied field $\mu_0 H = 9$ T. The data are normalized to that for the pure sample.

where $\sigma_{h(e)}$ is the conductivity of the hole (electron) band and $\mu_{h(e)}$ is the respective mobility. In view of the results in Fig. 3, the above expression ensures that the mobilities for holes and electrons are approximately the same above T_{SDW}. The situation is quite distinct in the ordered phase, where the MR is fairly large and positive. In this region Eq. (1) does not describe adequately the observed results. At first, as shown in Fig. 2, the experimentally determined MR may be described as $\Delta\rho \propto a\mathrm{H}^b$, where a is a constant and $b \approx 1.5$ for all samples and temperatures. This behavior deviates from the quadratic field dependence predicted by Eq. (1). Secondly, within the two-bands scenario one would have to suppose that some severe Fermi surface reconstruction at $T = T_{\mathrm{SDW}}$ [20,22] forces the mobilities μ_e and μ_h to become significantly different at this temperature. Moreover, this difference should increase as the temperature decreases into the magnetically ordered state.

We propose a description of the MR data in the temperature range below T_{SDW} by considering the effect of spin-dependent scattering. Since the 122 Fe-pnictides order antiferromagnetically, the application of a magnetic field disrupts the imbalance of the staggered magnetizations. Then, instead of reducing spin-disorder, as is the case in ferromagnetic metals, the field enhances the cross-section for spin-flip scattering. Roughly, one would expect that the field-dependent resistivity increases as [28]:

$$\rho(\mathrm{H}) \propto c\langle S\rangle^2, \tag{2}$$

where c is a constant and $\langle S\rangle \sim [n(\uparrow) - n(\downarrow)]$ is the field induced difference between the densities for electrons with spin parallel and anti-parallel to the field orientation if one assumes that the antiferromagnetism comes from a SDW state. On the other hand, $\langle S\rangle$ must be interpreted as the net magnetization if one supposes that localized moments in the Fe atoms governs the magnetically ordered phase of the studied compounds. In any case, the field disturbs the cooperative spin alignment and should increase the electron scattering rate. The fact that the MR increases as a power law of the field with exponent $b \sim 1.5$ indicates that $\langle S\rangle$ increases sublinearly as a function of H.

The occurrence of a magnetic contribution to the resistivity in our samples is also suggested by results plotted in Fig. 3. In these plots, one observes that the amplitude of the MR as a function of T is qualitatively reminiscent of an order parameter that becomes non-zero in temperatures $T < T_{\mathrm{SDW}}$. This behavior is independent of the applied field and is consistent with Eq. (2). We are thus led to consider that the MR of lightly substituted 122 Fe-pnictides is originated from dissipative electron scattering by spin excitations in the antiferromagnetic phase. This hypothesis contrasts with the previously proposed mechanisms based only on the two-band conduction model with large dominance of one type of carrier below T_{SDW} [20,21,24].

Fig. 4 shows $\Delta\rho = \rho(\mathrm{H}) - \rho(0)$ for our samples in temperatures $T = 20$ K and $T = 100$ K, and field $\mu_0\mathrm{H} = 9$ T. The data are normalized to that of the pure sample. The results in Fig. 4 roughly indicate that the general behavior of field-dependent electron scattering rate does not change drastically with chemical substitution and, at least at the concentration range studied here, it does not show a systematic dependence on the substituting atom.

Fig. 5 shows the Kohler plot for our Cu-substituted sample. The Kohler's rule is violated in a large temperature interval inside the magnetically ordered phase ($50\ \mathrm{K} < T < T_{\mathrm{SDW}}$). The Kohler's rule is generally invalid in multiple band conductors. Moreover, violations of Kohler's rule can be related to distinct scattering times of the charge carriers, with possible different temperature dependencies in the presence and absence of magnetic field and/or field dependent scattering times [29]. Thus, to our understanding, violations of Kohler's rule in our samples should be interpreted as an indication of the presence of the carrier scattering by magnetic fluctuations in the ordered phase of our samples.

[ht]

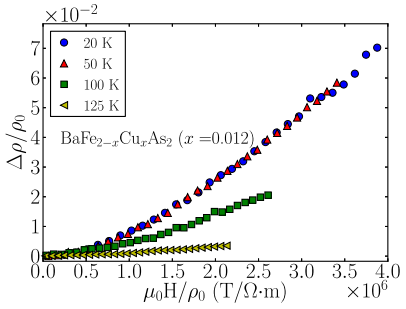

Fig. 5. Representative plot showing the violation of the Kohler's rule in the magnetic phase of the Cu-substituted sample for temperatures $T \gtrsim 50$ K.

3.3. Hall effect

Figs. 6 and 7 show the results from the Hall effect experiments carried out in the studied samples. In Fig. 6 the Hall resistivity ρ_{xy} is presented as a function of the magnetic field at several fixed temperatures. In all cases ρ_{xy} is a linear function of μ_0H. In this respect, our pure sample differs from others reported in literature [17] where the ρ_{xy} vs. μ_0H curves present a slight positive curvature for fields $\mu_0 H \gtrsim 2$ T. We speculate that our pure specimen presents some kind of disorder which has the same effect in ρ_{xy} as the addition of impurities. This could also explain the fact that our pure sample to have a $T_{SDW} \sim 135$ K, slightly lower than the commonly reported value $T_{SDW} \sim 140$ K. In Fig. 7 the Hall coefficient R_H is shown as a function of the temperature for all samples. The Hall coefficient was obtained from the slope of the straight lines fitted to the ρ_{xy} vs. μ_0H data in Fig. 6.

In Fig. 7 one observes that R_H is negative, small and weakly temperature dependent in the paramagnetic region ($T > T_{SDW}$). At the magnetic ordering temperature, a remarkable change occurs in the Hall coefficient: while remaining negative, R_H becomes strongly temperature dependent, so that its magnitude increases roughly linearly by more than one order of magnitude as T decreases toward zero. The negative sign of R_H in the whole studied temperature range indicates that, at these doping levels, the transport is dominated by electrons in all the studied samples. The fact that $|R_H(T)|$ decreases almost linearly to small values when T approaches T_{SDW} from below, and shows a remarkable change of behavior at this temperature, is in agreement with the temperature dependence of the magnetoresistance amplitude shown in Fig. 3. Also, the weak dependence of the R_H magnitude on the imputity atom is remarkable in the results of Fig. 7. Consequently, in accordance with the resistivity and magnetoresistance results, the Hall effect data in Fig. 7 suggest that substituting atoms at the Fe site of the 122 Fe-pnictides act mostly as scattering centers than as effective dopants, at least within the concentration limit studied here. This result is in agreement with previous reports which suggested that the main role of the distinct chemical substitutions in the Ba122 family is to provide local distortions near the Fe site [14,30,31], so that subtle variations of the structural parameters play a major role to explain the phyical properties of the Fe-pnictides [32].

The behavior of R_H in the paramagnetic region is in accordance with the expectations for conduction by two currents, consistently with the previously described magnetoresistance results. According to the two-band conduction model the Hall coefficient is written as [27]:

$$R_H \approx \frac{\sigma_h \mu_h - \sigma_e \mu_e}{(\sigma_h + \sigma_e)^2}. \quad (3)$$

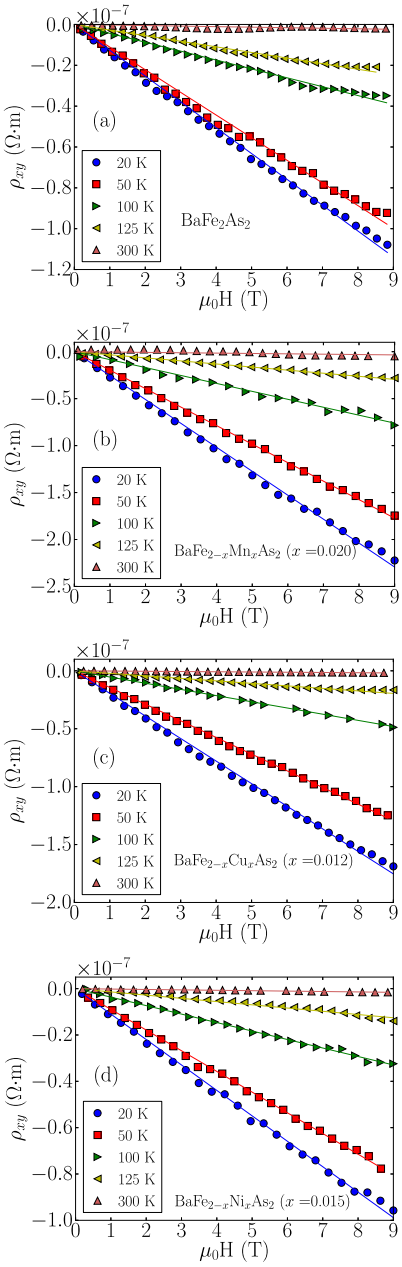

Fig. 6. Hall resistivity as a function of the applied magnetic field in several fixed temperatures. Solid lines are fit to straight lines.

The small values of R_H in the paramagnetic regime indicate nearly compensated transport by holes and electrons; it means that not only mobilities, but the electron and hole conductivities are close to each other in the paramagnetic phase in all cases.

In the paramagnetic region, where the effect of magnetic order can be neglected, authors in Ref. [33] theoretically proposed that the variation of R_H with the temperature can be understood by introducing an additional term to the Hall effect which origin is beyond the Boltzmann approximation. This supplementary term corresponds to vertex corrections to the quasi-particle currents. These corrections are argued to be necessary because of the mixing produced by the exchange of spin fluctuations between the electron (e) and hole (h) bands. Others suggest that the temperature dependence of R_H in $T > T_{SDW}$ is associated with the momentum-dependent scattering off spin fluctuations and the ellipticity of the electron pockets [34,35].

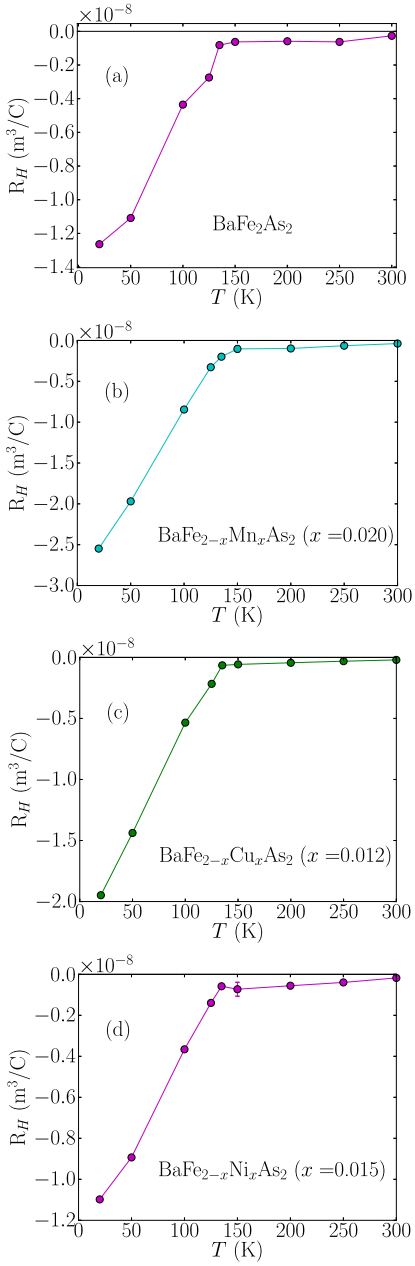

Fig. 7. Hall coefficient as a function of the temperature. The points represent the R_H value for each temperature was obtained from the slope of the straight line fits in Fig. 6.

On the other hand, explanations for the temperature dependence of R_H in the magnetically ordered region include, in most cases, a severe Fermi surface reconstruction generated by the orthorhombic distortion accompanied by the SDW ordering. While some authors consider the reduction of the charge carriers density [20,32,36], others propose that a reduction of the hole mobility or an enhancement of electron mobility is the dominant phenomenon [21,24].

Here, we propose that contributions related to magnetic excitations are also important to explain the Hall effect in the ordered state of 122 Fe-pnictides. In particular, we associate an extraordinary, or anomalous contribution to the Hall effect in addition to the ordinary one described by Eq. (3). Since the ordinary contribution is very small due to the almost compensated two-band conduction, we suggest that the R_H magnitude in the temperature region below T_{SDW} largely comes from the anomalous term.

The anomalous contribution to the Hall effect (AHE) is mostly associated with ferromagnetic metals. It is parametrized as a linear function of the magnetization, so that the total Hall resistivity is written as [37]:

$$\rho_{xy}(\mathrm{H}) = \mu_0(R_0\mathrm{H} + R_S\mathrm{M}), \tag{4}$$

where μ_0 is the vacuum permeability, R_0 is the ordinary Hall coefficient, M is the magnetization and R_S is the anomalous Hall coefficient. The first term in the right side of Eq. (4) represents the ordinary Hall effect due to the Lorentz force and the second one is the anomalous contribution. At first glance, since antiferromagnetic metals have zero net spontaneous magnetization, Eq. (4) rules out the description of the Hall effect in these materials. Consequently, up to now an anomalous contribution to the Hall effect has not been taken into account for explaining the behavior of the magneto-transport phenomena in Fe-pnictides. However, we must note that antiferromagnetic metals can also develop an AHE induced by non-collinear spin structures [38,39], and even non-frustrated collinear lattices may develop anomalous contributions to the Hall current [40] as observed in the U_2PdGa_3 [41] and $Nd_{1-x}Ca_xB_6$ [42] compounds.

Equation (4) is an empirical relation which should not be taken as universal, neither suitable for all materials [37]. However, since antiferromagnetic materials can develop a field-induced magnetization, which may be approximately written as $\mathrm{M} = \chi_{eff}(T)\mathrm{H}$, where $\chi_{eff}(T)$ is a temperature dependent effective susceptibility, here we assume the validity of Eq. (4) to describe the results in Figs. 6 and 7. Then we write:

$$\rho_{xy}(\mathrm{H}) = \mu_0 R_H \mathrm{H}, \tag{5}$$

where

$$R_H = R_0 + \chi_{eff}(T)R_S(T). \tag{6}$$

Based on the results shown in Fig. 7, we suppose that $R_H \simeq R_0$ in the paramagnetic phase. A small contribution from an anomalous term coming from scattering by magnetic impurities, and corrections as those considered in Refs. [33–35], might lead to the temperature dependence of R_H in the $T > T_{SDW}$ region. As a working hypothesis, we assume that R_0 remains small ($R_0 \sim 10^{-9}$ C/m^3 in all cases) below T_{SDW}, so that the strong temperature dependence of R_H in this temperature range is due to the anomalous Hall contribution. Thus, we suppose that $R_H(T) \simeq \chi_{eff}(T)R_S(T)$ for $T < T_{SDW}$.

Key ingredients to develop AHE are the multi-orbital character of the charge carriers, spin-orbit interaction and time-reversal symmetry breaking. The multi-orbital character of the carriers in the Fe-pnictides is already known. The existence of spin-orbit interaction in these compounds was theoretically predicted [43], and experimentally corroborated by ARPES measurements [44]. The application of a magnetic field naturally breaks the time-reversal-symmetry, but an intrinsically broken time-reversal symmetry related to the particular magnetic ordering in the 122 compounds should be present in order to generate an enhanced AHE. At this point, one might consider that some “hidden” magnetic order [45] accompanying the SDW state plays a role to explain the broken time-reversal symmetry in the 122 Fe-pnictides.

Taking the above considerations into account, we propose that the inclusion of an anomalous term for explaining the Hall effect results in the magentic state of our samples is a reasonable assumption. Of course this statement leads to the issue of determining the origin of the AHE in these materials. Four mechanisms are conventionally assumed to produce AHE in ferromagnetic materials [37,46]. The so-called intrinsic mechanisms are related to Berry phase effects on the Bloch wave-functions of the charge carriers

under the influence of spin-orbit scattering. Two different possibilities may occur: the Berry phase may accumulate in the reciprocal space giving origin to a dissipationless transverse current [47], or the carriers can accumulate a Berry phase in the real space due to canting of localized spins. The former is called the Karplus–Luttinger (K-L) contribution [47] and the last one is known as the spin-chirality mechanism [48]. Two extrinsic mechanisms for AHE are due to the interaction of the charge carriers with atomic magnetic scattering centers in the material and depend on the concentration of those single-ion moments; these are the skew-scattering and side-jump contributions [46].

Identifying experimentally the contribution of each mechanism to the AHE of a given material may be difficult. In general, a useful procedure is to plot the anomalous Hall coefficient as a function of the longitudinal resistivity assuming that these quantities relate to each other through a simple power-law with the form $R_S \propto \rho_{xx}^{\beta}$. The K-L theory predicts that $\beta = 2$ [46,47]. For the skew-scattering mechanism the exponent is $\beta = 1$ provided that ρ_{xx} increases linearly with the concentration of single-ion moments. For the side-jump one also expects $\beta = 2$, since the effect should depend on the square of the single-ion moment concentration [46]. As a general trend, the K-L mechanism is expected to describe the AHE in systems where the magnetic moments distribute periodically in the lattice. Skew-scattering is dominant in dilute magnetic alloys while side-jump should prevail in concentrated magnetic alloys. In the case of our experiments, since $\chi_{eff}(T)$ data are not available for our samples, we are unable to single out the value of $R_S(T)$ from the R_H value. However, measurements of M/H existing in the literature for pure and substituted 122 compounds, with similar and higher content of the Mn impurity [49] show a temperature dependence that is reminicent of the R_H results in Fig. 7(b). In other words, the temperature dependence of R_H can be largely dictated by $\chi_{eff}(T)$.

Even so, if we consider that $\chi_{eff}(T)$ is approximately constant in a large temperature interval inside the magnetic region in pure and very low substituted samples [49,50], we obtain the relation $R_H \propto R_S(T) \propto \rho_{xx}^{\beta}$. Thus, in this case we can evaluate the dependence of R_H, or equivalently of ρ_{xy}, on ρ_{xx}. The results for ρ_{xx} in Fig. 1 and ρ_{xy} in Fig. 6 show that the longitudinal and Hall resistivities in the samples studied here are in anti-correlation in the magnetically ordered phase; that is, ρ_{xx} increases while ρ_{xy} decreases when plotted as functions of the temperature in the range $T < T_{SDW}$. Then, one can not relate the Hall resistivity to a single power-law of the longitudinal resistivity with exponent $\beta = 1$ or $\beta = 2$. This fact suggest that the AHE in our samples could be due to the spin-chirality mechanism [51]. The theory for this effect does not predict any correlation of R_S with the longitudinal resistivity [52]. When identified experimentally, the contribution of the spin-chirality to the AHE does not show any obvious dependence with $\rho_{xx}(T)$ [53]. The spin-chirality mechanism implies that canting of local spins with respect to the magnetization must occur, so that the triple product $(\vec{S_i} \bullet \vec{S_j} \times \vec{S_k})$ of neighboring spins is non-zero. This canting may be static and related to local disorder. Because of the presence of microscopical defects as twinning, dislocations and vacancies, lattice distortions are likely to occur in profusion even in single crystal samples of the 122 Fe-pnictides (especially in the presence of chemical substitution) [41]; consequently, spin-chiralities can be good possible generators of the AHE in this systems. However, the mechanism of spin-chiralities can also be relevant in collinear spin systems, provided that the inelastic scattering rate for conduction electrons is larger than the relaxation rate for the spin excitations [53].

In order to obtain an additional insight on the Hall effect in the low temperature region, in Fig. 8 we plot the tangent of the Hall angle ($\tan\Theta_H = \rho_{xy}/\rho_{xx}$) as a function of the temperature for the

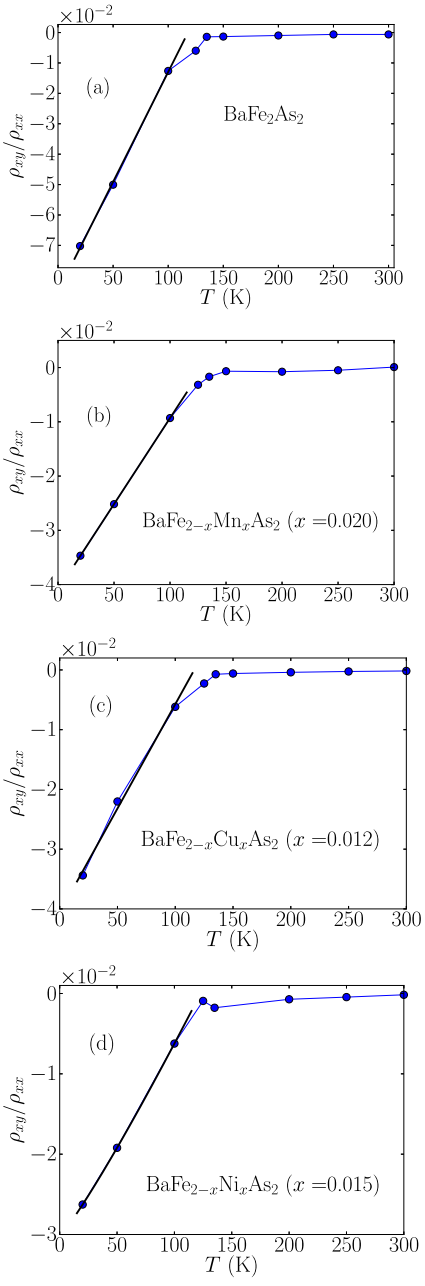

Fig. 8. Tangent of the Hall angle for an applied field $\mu_0 H = 4$ T as a function of the temperature for the studied samples. Solid straight lines in the low temperature region are fits of the experimental points to Eq. (7).

studied samples. This parametrization for the Hall effect is of limited value here since the residual resistivity represents a large contribution to the total resistivity of these samples in the whole temperature range. However, a remarkably simple result is obtained in the region $T < T_{SDW}$, where $\tan\Theta_H$ behaves as a linear function of the temperature as:

$$\tan\Theta_H = \alpha - \beta T. \tag{7}$$

Table 2 lists the parameters α and β obtained from the best fits to Eq. (7) in the low temperature range in Fig. 8.

The behavior of the tangent of the Hall angle in the magnetically ordered region as described by Eq. (7), can not be explained with the single conduction band model. In this case, one expects that $\tan\Theta_H = \omega_c\tau$, where ω_c is the cyclotron frequency and τ is the electron transport relaxation time. In the simplest two-

Table 2
Fitting parameters for the tangent of the Hall angle in the magnetic phase of the listed systems according to Eq. (7).

Sample	$-\alpha(10^{-2})$	$\beta(10^{-4})$ (K^{-1})
$x = 0$	8.5	7.2
$Mn_{0.020 \pm 0.006}$	4.1	3.2
$Cu_{0.012 \pm 0.004}$	4.1	3.5
$Ni_{0.015 \pm 0.005}$	3.2	2.5

band model, one may write $\tan\Theta_H = (\sigma_h\omega_h\tau_h - \sigma_e\omega_e\tau_e)/(\sigma_h + \sigma_e)$, where $\omega_{h(e)}$ and $\tau_{h(e)}$ are the cyclotron frequencies and relaxation times for holes (electrons), respectively. Unless assuming a rather unusual and somewhat conflicting temperature dependencies for the relaxation times and/or conductivities for holes and electrons, it is impossible to describe results for $\tan\Theta_H$ in the region $T < T_{SDW}$ with basis solely on two-band conduction. Then, results in Fig. 8 also led us to suppose that the Hall effect in the magnetically ordered state of the undoped and slightly doped 122 Fe-pnictides is mostly due to an anomalous component. This component behaves differently than those identified in ferromagnetic systems and should be related to the particular antiferromagnetic ordering of the 122 Fe-pnictides, where both local moment and SDW type orderings coexist [5–7]. We note that ρ_{xy} and ρ_{xx} are also in anti-correlation in the cuprate superconductors (HTSC), so that the tangent of the Hall angle in these materials also decreases when the temperature increases [54,55]. As for the Fe-pnictides, the existence of AHE in the HTSC is a matter of controversy. However, the presence of antiferromagnetic correlations is relevant in these two systems. In the pure and slightly doped Fe-pnictides, long-range SDW stabilizes into an antiferromagnetic ordering at non-zero temperatures. By using quasi-particle interference imaging, antiferromagnetic spin fluctuations have already been identified as the principal electron-boson interaction in Fe-pnictides [56]. In the HTSC, spin fluctuations have been detected by neutron scattering experiments and may underly the pseudogap phenomenon [57]. These excitations may be short-ranged and dynamical SDW, but can significantly affect the transport properties if they last long enough in comparison to the carrier relaxation time.

It is also relevant to observe that the temperature dependence of the Hall angle shown in Fig. 8 and described by Eq. (7) extrapolates to a non-zero and large value at $T = 0$. Within the interpretation that ascribes a magnetic origin for ρ_{xy} at low temperatures, this result also suggests that the role of spin chiralities is dominant to explain the Hall effect in the 122 Fe-pnictides.

As a final remark, we stress that the multi-band character of the charge carriers in the studied compounds must also be taken into consideration in order to completely describe their magneto-transport properties. Two-band conduction explains the small magnetoresistance and Hall resistivity generally observed in the paramagnetic high temperature phase. Although authors in Ref. [58] consider that three bands, two electron-type and one hole-type, must be considered to fully account for the magneto-transport properties of the pure $BaFe_2As_2$ compound, in the phenomenological description of the magnetoresistance and Hall effect results in our substituted compounds, it seems good enough to consider a simpler picture where a single current of electron-type carriers adds to conduction by holes. Our results indicate that multi-band conduction alone hardly explains the behavior of both properties in the low-temperature ordered phase, since one has to suppose that besides the occurrence of a drastic Fermi surface reconstruction at T_{SDW}, progressive modifications of the band structure ocurr below this temperature. A physically more simple description of the magneto-transport properties of the 122 Fe pnictides might be achieved by considering the effect of scattering by magnetic excitations, which have been proven to be important in these compounds [43,44,56].

4. Summary

We have measured the electrical resistivity, magnetoresistance and Hall effect in slightly substituted samples of the $BaFe_{2_x}TM_xAs_2$ system. Four single crystals were investigated, one pure sample with no Fe substitution, and three samples where TM = Mn, Cu, and Ni. Within the studied concentration regime, all three types of substituting atoms diminish the magnetic temperature transition by approximately 15 K with respect to that of the pure sample. From our resistivity and magneto-transport experiments we conclude that in the low dilution limit the chemical substitutions at the Fe sites rather behave as scattering centers, and little changes are produced in the carrier density.

The obtained results were discussed with basis on a scenario where the magneto-transport phenomena in the magnetically ordered phase of 122 Fe-pnictides are mostly governed by magnetic excitations. We then assume the occurrence of magnetic, or anomalous, contributions to the magnetoresistance and to the Hall effect in the ordered state. Indications of the validity of our approach are: (i) the temperature dependence of the magnetoresistance amplitude is reminiscent of a magnetic order parameter which becomes non-zero at $T \leq T_{SDW}$; (ii) a large and temperature dependent anomalous Hall contribution explains the enormous increase in the absolute value of the total Hall coefficient in the magnetic phase; (iii) the anomalous Hall resistivity does not show a simple power law scaling with the longitudinal resistivity; (iv) the tangent of the Hall angle varies linearly with the temperature below T_{SDW} and extrapolates to a finite value at $T = 0$,suggesting that the anomalous Hall effect of the 122 Fe-pnictides is due to the spin-chirality mechanism.

Finally, the multi-orbital character of the charge carriers and some Fermi surface reconstruction at the structural and magnetic transition might be taken into account in a detailed description of the magneto-transport properties of the studied compounds. In particular, two-band conduction is the most natural explanation for the practically zero magnetoresistance and the very small Hall resistivity that are systematically observed in the paramagnetic high temperature phase. In the magnetic ordered phase, however, scattering by magnetic excitations plays a relevant role.

Acknowledgments

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