Extraordinary Hall effect in Fe-Cr giant magnetoresistive multilayers

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We show that for giant magnetoresistive Fe-Cr multilayer samples, the scaling law for the extraordinary Hall constant, $R_S \sim \rho^n$ gives unrealistically large values of $n(n=2.8–3.3)$ if we use the resistivity ρ at an external field $B=0$ as done by previous investigators. With B perpendicular to the film plane, the transverse magnetoresistance saturates at $B \approx 3$ T in the 4.2–300 K temperature range. This indicates that the Fe layers, although antiferromagnetically coupled at $B=0$, are ferromagnetically aligned only above $B \approx 3$ T. Therefore, we use $\rho=\rho(B=3\text{ T})$ in $R_S \sim \rho^n$ and obtain $n=1.96\pm0.02$, which implies that the side-jump mechanism $n=2$ is the dominant scattering process. We emphasize that our interpretation of $R_S(\rho)$ not only evolves from a proper understanding of the magnetic state of the multilayer system but also yields physically meaningful values of the exponent n.

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I. INTRODUCTION

The Hall resistivity in ferromagnetic metals and alloys in a polycrystalline form is given by[1]

$$\rho_H = R_0 B + \mu_0 R_S M_S, \tag{1}$$

where R_0 is the ordinary Hall constant (OHC) R_S is the extraordinary or spontaneous Hall constant (EHC), B is the magnetic induction, M_S is the saturation magnetization, and μ_0 is the permeability of the free space. The first term is due to the Lorentz force acting on the charge carriers and is present in nonmagnetic materials as well. The second term is a characteristic of the magnetic state of the material and is proportional to the magnetization. Two different mechanisms are responsible for R_S. They are the classical asymmetric scattering and the nonclassical "side-jump" mechanism, both having their origin in the spin-orbit interaction present in a ferromagnet. R_s satisfies a power law for both of them, given by

$$R_s = a\rho + b\rho^2, \tag{2}$$

where the linear and quadratic terms in ρ are due to asymmetric scattering and side-jump mechanisms, respectively,[1] and ρ is the resistivity at $B=0$.

The classical Boltzmann equation is correct to lowest order in the dimensionless parameter $\hbar/\tau E_F$, where τ is the relaxation time and E_F is the Fermi energy. For pure metals and dilute alloys at low temperatures $\hbar/\tau E_F \ll 1$. Smit first introduced the idea of asymmetric or skew scattering of electrons by impurities. In the presence of spin-orbit interaction (between the spin of the magnetic electron and the orbital angular momentum of the scattered conduction electron) in a ferromagnet, there is a left-right asymmetry in the differential scattering cross section about the plane defined by $\boldsymbol{J}$ and $\boldsymbol{M}$,[2] $\boldsymbol{J}$ being the current density. As a result, electrons tend to pile up on one side of the sample, giving rise to a transverse voltage. A net polarization results because of ferromagnetism, i.e., different densities of states of the up- and down-spin bands at the Fermi level due to exchange splitting. Using the classical Boltzmann equation in the relaxation time approximation, one calculates ρ_H using the boundary condition $J_y=0$, and finds that $R_s=a\rho$. In the other limit ($\hbar/\tau E_F$ is not very small), nonclassical transport is expected to dominate as in concentrated alloys and at high temperatures. Earlier nonclassical theories predicted $R_s \sim \rho^2$, in agreement with experiments, but they are rather complicated. A simple intuitive theory was given by Berger,[3] which suggested a physical mechanism responsible for the Hall effect in ferromagnets. It is assumed that a conduction electron is scattered by a central impurity potential and travels in straight lines before and after scattering. In the presence of spin-orbit interaction, the symmetry of the problem is low and the two straight lines may not coincide. Either they make an angle δ, related to the asymmetric scattering or the two lines do not meet at the scattering center and give rise to a small but abrupt side-jump ΔY. Including spin-orbit interaction due to both impurity and periodic potentials, ΔY is calculated to be ≈ 1 Å. Finally a transport theory for arbitrary $\omega_c\tau$ (ω_c is the cyclotron frequency) yields Eq. (1) with $R_S=b\rho^2$.

The field and temperature dependence of the Hall effect and the magnetoresistance in giant magnetoresistive (GMR) magnetic multilayers like Fe-Cr should shed new light on their magnetic states and scattering processes. Several studies on the Hall effect have been reported, but detailed temperature-dependence studies are only a few. Aoki *et al.*[4] studied the Cr-layer thickness (d_{Cr}) dependence of ρ_H and the giant magnetoresistance (GMR) in magnetron sputtered Fe-Cr multilayers. $\rho_H(B)$ behaved like those of ordinary fer-

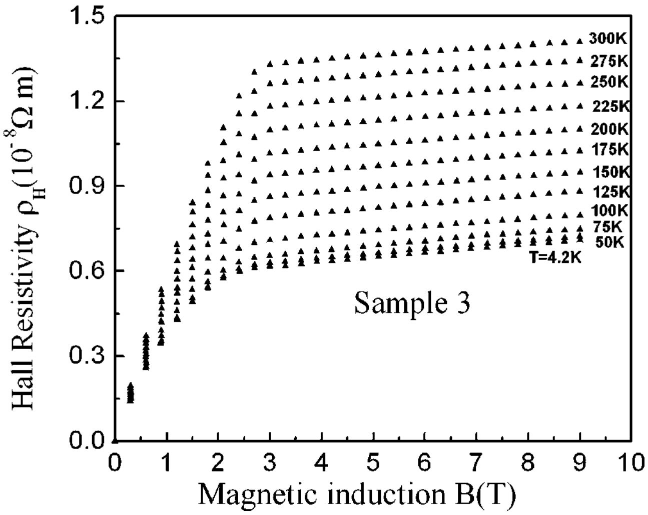

FIG. 1. Hall resistivity (ρ_H) is plotted against magnetic induction B up to 9 T at several temperatures from 4.2 to 300 K for sample 3. They show the typical behavior of a ferromagnet at high fields. In the Hall geometry $\mathbf{B}=\mu_0\mathbf{H}_{applied}$ since the demagnetization factor $N\approx 1$.

romagnetic films with the increase of d_{Cr}, whereas it had shown an anomalous behavior for smaller d_{Cr} where the GMR was, however, the largest. R_0 was found to be positive increasing with d_{Cr}. Also, they found a negative asymmetric scattering term as well as a positive (weakly d_{Cr}-dependent) side-jump contribution. However, all the measurements were done only at 4.2 K. The effect of annealing temperature on the Hall effect was studied by Lucinski *et al.*[5] in dc magnetron sputtered Fe-Cr multilayers. A strong correlation was found between the GMR, R_S, and M_S, all showing maxima around the annealing temperature of 270 °C where structural defects may have been annihilated. A comprehensive study of the surface roughness dependence of the Hall effect were reported by Korenivski *et al.*[6] on Fe-Cr multilayers grown on Si substrates using dc magnetron sputtering. The surface roughness was changed by varying the sputtering gas pressure and by isothermal annealing. Samples at the lowest argon pressure of 4 mTorr had shown the best antiferromagnetic state ($M\approx 0$) at $B=0$. The exponent, n, of ρ of Eq. (2) increased from 2.0 to 2.3 as the argon pressure was increased, which in turn, increased the interfacial roughness. Low-temperature annealing increased the roughness and enhanced R_S and n. R_0 was found to be roughly independent of temperature below 250 K, and the argon gas pressure and annealing hardly affected it.

It is interesting to compare the behavior of R_S in bulk Fe and its alloys[1] with those in the Fe-Cr multilayers. At low temperatures, in pure Fe and Fe-Co dilute alloys, R_S is negative and is dominated by asymmetric scattering ($R_S\sim\rho$, $n=1$) whereas for Fe-Cr and other concentrated alloys it is the side-jump mechanism ($R_S\sim\rho^2$, $n=2$) which dominates. In case of magnetic multilayers, as summarized in Ref. 6, $n=2$ for molecular beam epitaxy grown Co-Cu superlattices, $n=2.6$ for *e*-beam evaporated Fe-Cr multilayers, and $n=3.7$ for Co-Ag films. Thus these inhomogeneous materials behave quite differently from bulk Fe and its alloys. Zhang had shown,[7] for magnetic multilayers, the failure of the scaling law, i.e., Eq. (2), which generally holds for homogeneous magnetic materials. The actual functional dependence is governed by scattering parameters in the bulk and at the interfaces and also on the spin-orbit coupling.

Here we report the high-field Hall resistivity and transverse magnetoresistance data in some ion-beam-sputtered Fe-Cr multilayers with magnetic structures, which depend strongly on external magnetic fields. These multilayers had shown[8] a longitudinal negative giant magnetoresistance of $\approx$21% at 4.2 K, saturating at $\approx$1.3 Tesla. The magnetic field $\boldsymbol{B}$ and the current $\boldsymbol{J}$ were in the film plane with the current parallel to the field $\boldsymbol{J}\parallel\boldsymbol{B}$. The GMR could be understood in terms of the spin-dependent bulk scattering in ferromagnetic Fe layers. The ferromagnetic Fe layers, in zero or small magnetic fields, are antiferromagnetically coupled through the Cr spacer layers via the oscillatory Ruderman-Kittel-Kasuya-Yosida interaction mediated by the conduction electrons. Here a spin-dependent scattering results in a high electrical resistance. An external magnetic field gradually aligns the spins in different Fe layers and produces a completely ferromagnetic arrangement beyond a field H_{sat}. The majority band electrons in bulk Fe have large conductivity and they produce some kind of short-circuiting effect causing a decrease in resistance and hence a large negative GMR. Interface scattering also contributes to the GMR.

II. EXPERIMENTAL DETAILS

Fe-Cr multilayers of composition Si/Cr(50 Å)/[Fe(20 Å) / Cr(t(Å))]30 / Cr((50$-t$(Å)) where $t=10$–12 Å ($t=10$ Å for sample 1 and 12 Å for samples 2 and 3, respectively) were grown[8] at MCNC, North Carolina, on a Si substrate by sputtering with Xe ions at 900 V and a beam current of 20 mA. Samples 2 and 3 were deposited under slightly different base pressure. The Hall resistivity was measured at the Indian Institute of Technology, Kanpur, using a five-probe dc method in order to minimize the resistive voltage arising from the misalignment of the Hall voltage probes. A voltage divider was made using two General Radio decade resistance boxes (Type-1432-M and -B). A current of 10 mA was passed through the sample ($\approx 10\times 3\times 0.0001$)$mm^3$ from a Keithley 224 programmable current source. The Hall and magnetoresistive voltages were measured by a Prema 6001 digital multimeter. The misalignment voltage was adjusted to within 0.1 μV or better at each temperature. The temperature was controlled and measured within 0.1 K by a Lakeshore 340 temperature controller and a calibrated CERNOX (Model CX-1030-SD)thermometer. A 9-Tesla superconducting magnet with Intermagnetics power supply (Model 150-M)provided the necessary magnetic field. The Hall voltage (typically 300 μV)was measured with an accuracy of 1 part in 3000 whereas the resistive voltage was accurate to within one part in 10^5. The sign of the Hall voltage was found to be positive using the Lorentz force law as well as by a comparison with an iron sample having a positive Hall voltage at 300 K.

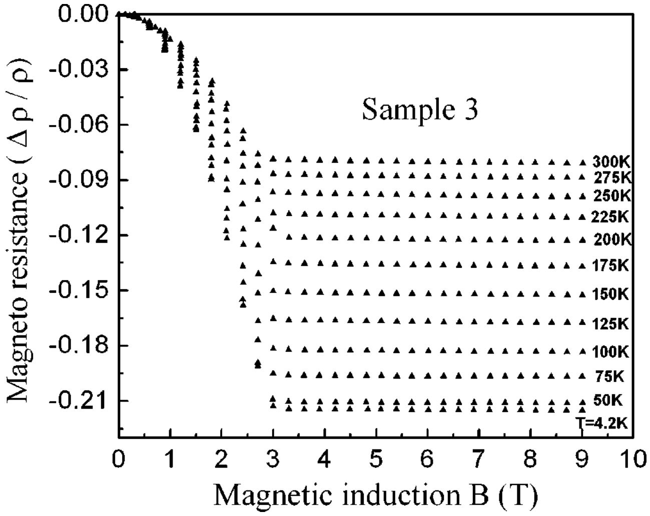

FIG. 2. Transverse magnetoresistance ($[\rho(B)-\rho(0)]/\rho(0)$) is plotted against magnetic induction B up to 9 T at several temperatures from 4.2 to 300 K for sample 3. The negative transverse MR is $\approx 21\%$ at 4.2 K, the same as in the longitudinal case except $H_{Sat}\approx 3$ T here as against 1.3 T in the longitudinal case (Ref. 6).

III. RESULTS AND DISCUSSION

In Fig. 1 we have plotted the Hall resistivity (ρ_H) vs magnetic induction (B) for sample 3 at several temperatures. In the Hall geometry the magnetic field is perpendicular to the film plane and so the demagnetization factor $N\approx 1$. Therefore, $\boldsymbol{B}=\mu_0[\boldsymbol{H}_{applied}+(1-N)\boldsymbol{M}_S]\cong\mu_0\boldsymbol{H}_{applied}$. ρ_H shows the typical behavior of a ferromagnet varying linearly with B at fields higher than 3 T, in accordance with Eq. (1). This is also borne out by Fig. 2 where the transverse magnetoresistance (MR)$=[\rho(B)-\rho(0)]/\rho(0)$ is plotted against B for sample 3 at several temperatures. It is clear that the MR saturates more or less at all temperatures only above 3 T

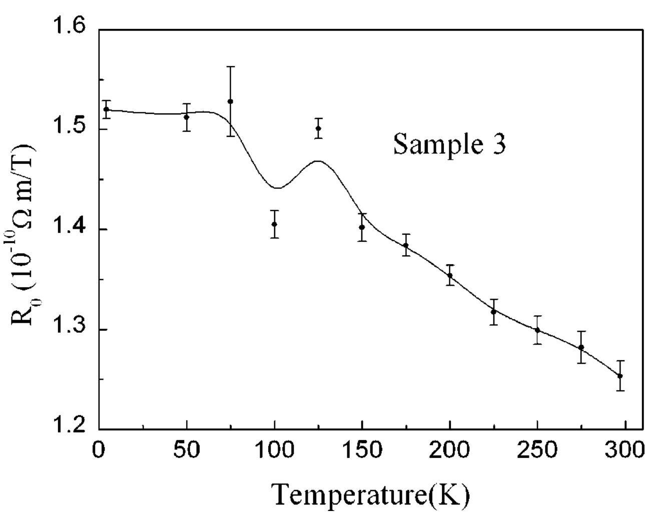

FIG. 3. Hall constant R_0 is plotted against T for sample 3. The solid line is just a guide to the eye. R_0 shows an overall marginal decrease with increasing temperature, indicating hardly any change in the conductivities of the electron and hole bands. The error bars of R_0 are also shown.

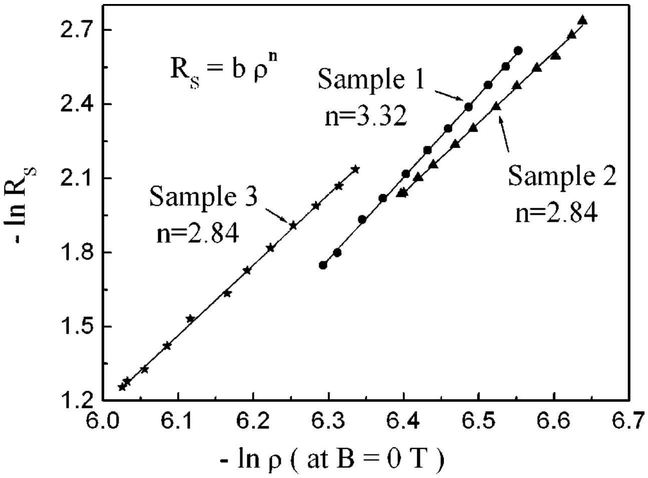

FIG. 4. $-\ln R_S$ is plotted against $-\ln\rho$ (ρ is the resistivity at $B=0$) at each temperature for finding n of Eq. (2) assuming $a=0$. From the linear fits the values of n are found to be 3.32, 2.84, and 2.84 for samples 1, 2, and 3, respectively.

indicating that the ferromagnetic alignment of the Fe layers in the direction of the field is complete at fields much higher than those for the longitudinal magnetoresistance reported earlier[8] where the MR saturated above only 1.3 T. However, the GMR ($\approx 21\%$) is the same in both the orientations, only the saturation fields are different (3 and 1.3 T for transverse and longitudinal MR, respectively). B is still ≈ 3 T for the longitudinal case as well, where $N=0$ and $\mu_0 M_S\approx 1.7$ T. The slope above 3 T gives R_0 and the intercept with the ρ_H axis gives $R_S M_S$. $M_S(T)$ was measured[9] using a Quantum Design superconducting quantum interference device in magnetic fields $>H_{Sat}$. Thus one gets $R_S(T)$.

In Fig. 3 we have shown R_0 vs. T for sample 3. The solid line is just a guide to the eye. The error bars, obtained from the fit to Eq. (1), are also shown. R_0 shows an overall decrease with increasing temperature. In a two-band model of a compensated ferromagnet (like Fe in these Fe-Cr multilay-

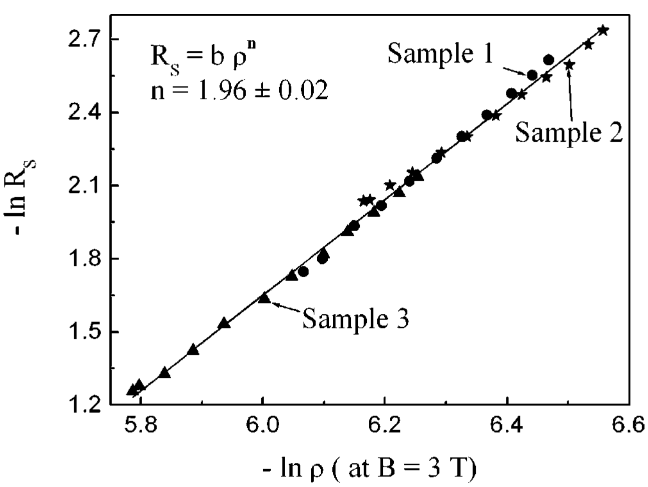

FIG. 5. $-\ln R_S$ is plotted against $-\ln\rho$ using ρ at $B=3$ T from Fig. 2. The data for all three samples fall on a common straight line with a slope of $n=1.96\pm 0.02$. The side-jump mechanism gives $n=2$ for homogeneous ferromagnets.

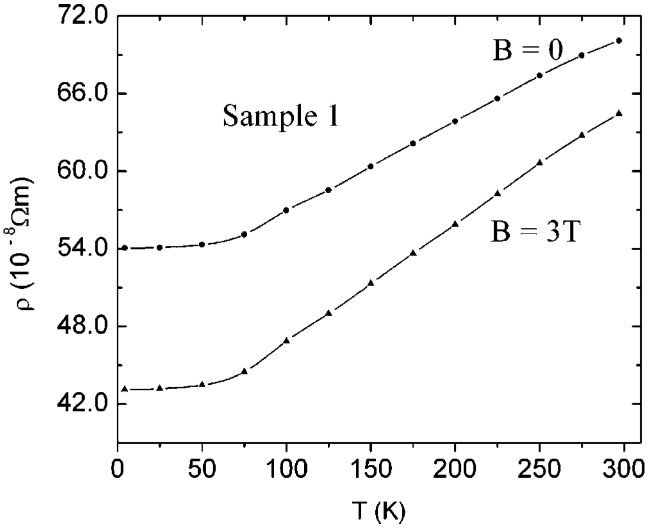

FIG. 6. Electrical resistivity(ρ) vs T for sample 1 at $B=0$ (antiferromagnetic state) and $B=3$ T (ferromagnetic state) in the transverse orientation. The solid lines are just guides to the eye.

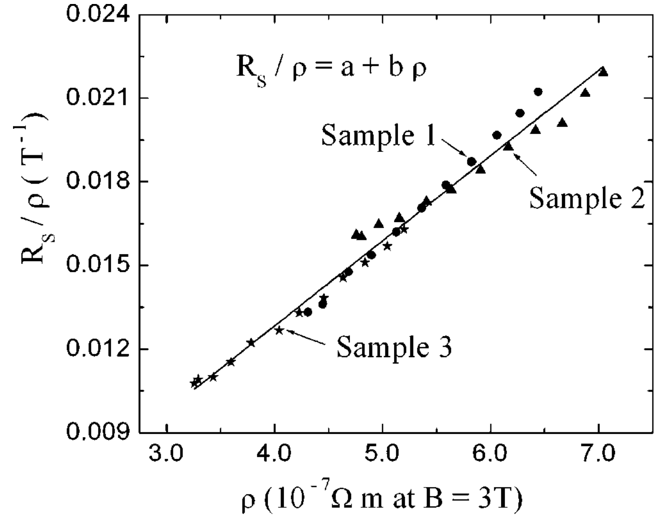

FIG. 7. R_S/ρ is plotted against ρ (points) for all the three samples where ρ is the resistivity at 3 T. The solid line is the best-fitted straight line whose slope gives $b=3\times10^4 m/\Omega^2 C$. The small positive intercept (a)$=(6\pm4)\times10^{-4}T^{-1}$ with the Y axis indicates the insignificant contribution of the asymmetric scattering in comparison to the side-jump mechanism.

ers), R_0 is proportional to the difference in conductivity between the hole and the electron bands[10]. Here (Fig. 3) the small positive value of R_0 indicates that the hole band is slightly more conducting than the electron band and remains so over a wide range of temperature (4.2–300 K) implying hardly any change in the band conductivities.

R_S, on the other hand, is found to increase with increasing temperature. The temperature dependence of $R_S M_S$ (increasing by a factor of 2.3 from 4.2 to 300 K) is mostly contributed by $R_S(T)$ since $M_S(T)$ decreases by only 6%. In order to find the ρ dependence of R_S [Eq. (2)] in Fig. 4 we have plotted [assuming a=0 in Eq. (2)] $-\ln R_S$ vs $-\ln\rho$, where ρ is the resistivity at $B=0$. We find that the data for the three samples follow different straight lines with slopes of $n=3.32$, 2.84, and 2.84, respectively for samples 1, 2, and 3. Such large values of $n(>2)$ have been found also in earlier work,[6] as mentioned in Sec. I. However, considering the fact that, at $B=0$, the Fe layers are antiferromagnetically coupled[8] while they are ferromagnetically aligned only beyond 3 T (see Fig. 2), in Fig. 5 we have plotted $-\ln R_S$ vs $-\ln\rho$ using ρ at $B=3$ T from the data shown in Fig. 2, and not at $B=0$. Now we observe that the data for all the three samples fall on the same straight line giving an excellent fit to Eq. (2) (taking $a=0$) with $b\approx2\times10^4 m/\Omega^2$C, slope of $n=1.96\pm0.02$, correlation coefficient of ≈0.996, and a small error in n. This value of n is very close to the prediction of the quantum transport theory where $R_S\sim\rho^2$, independent of the nature of the scatterer.[1] In Fig. 5, the variation of ρ is due to both impurity (ρ varying from 32 to 47 $\mu\Omega$ cm at 4.2 K in the three samples) and phonon scattering (4.2 to 300 K).

One might wonder how a 20% change of ρ at 4.2 K between 0 and 3 T can affect the exponent n so radically (from ~3 to ~2). This becomes clear from Fig. 6, where we plot ρ vs T at $B=0$ and 3 T for, say, sample 1. The fractional change in resistivity from 4.2 to 300 K $((\rho_{4.2K}-\rho_{300K})/\rho_{4.2K})$ is ≈0.297 at $B=0$ and 0.495 at $B=3$ T. On the other hand, the fractional change in R_S for sample 1 is as large as 1.315 from 4.2 to 300 K. Needless to say that a higher value of n (here 3.3) will fit the data at $B=0$ and a lower value of n (here 2.1) in the case of $B=3$ T. Thus we conclude that the use of ρ at $B=0$ is not justified since the multilayers are not even ferromagnetic at $B=0$ and Eqs. (1) and (2) do not apply in the antiferromagnetic state. Instead the use of ρ at 3 T, where the multilayers are completely ferromagnetically aligned, is justified. A further test of our new interpretation will be to perform similar analysis on a set of samples with wide variation of Cr thickness say, 6 – 20 Å, as against only 10 – 12 Å in our samples. In that

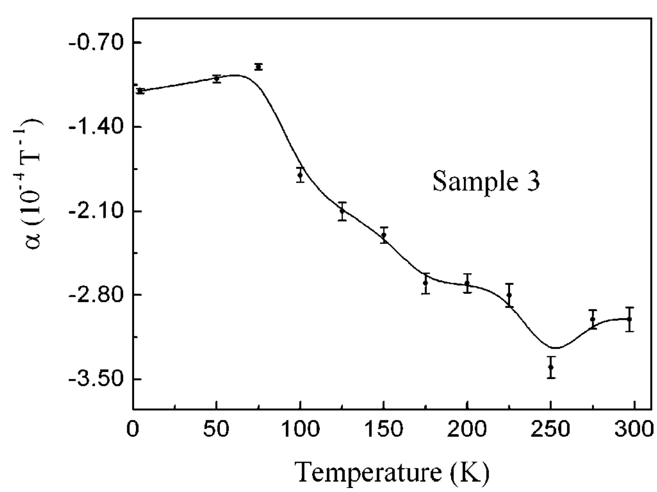

FIG. 8. $\alpha[1/\rho(\partial\rho/\partial B)]$ vs T for sample 3. α increases with increasing temperature implying less electron-magnon scattering at high temperatures. The error bars are also shown in the plot. The value of α at 300 K $(-3\times10^{-4}T^{-1})$ is comparable to the value $-4\times10^{-4}T^{-1}$ for bulk crystalline Fe.

case the zero field exchange coupling will vary from ferromagnetic to antiferromagnetic one giving rise to a large variation in the GMR. However, in a few cases,[6] values of n close to 2 were reported even by using ρ at $B=0$.

In general, both asymmetric scattering and nonclassical mechanisms could contribute to R_S. In order to find out the contribution from the former, we have plotted (points) R_S/ρ vs ρ [Eq. (2)] for all the samples in Fig. 7, again taking ρ at $B=3$ T. The slope of the best-fitted straight line (solid line in Fig. 7) gives the value of $b=(3.0\pm0.1)\times10^4 m/\Omega^2$C which is somewhat larger than that obtained from the $-\ln R_S$ vs $-\ln\rho$ plot (Fig. 5). The difference is due to the fact that the plot in Fig. 7 assumes $n=2$ whereas in Fig. 5 n is a parameter which is found to be ≈ 1.96. This value of b is comparable to $4\times10^4 m/\Omega^2$C found in dilute alloys of Fe with Cr impurities at 300 K.[1] Some mixing of Fe with Cr at the interfaces may be responsible for such similar values of b. It is interesting to note from the rather small positive intercept $a=(6\pm4)\times10^{-4}T^{-1}$ of the best-fitted straight line of Fig. 7 that the contribution to R_S from asymmetric scattering is not significant at all. The side-jump term $b\rho^2$ of Eq. (2) is the most dominant contributor to R_S.

Bulk ferromagnets (like Fe) at temperatures well below their Curie temperatures (T_C) show a negative magnetoresistance beyond technical saturation.[1] As the external magnetic field increases, it quenches the long-wavelength magnons because of the additional Zeeman term ($=g\mu_B B$, where g is the Lande g factor, μ_B is the Bohr magneton, and B is the external magnetic field) in the spin wave dispersion relation. As a result the electron-magnon scattering decreases with the increase of field and the electrical resistivity drops giving rise to a negative magnetoresistance. In view of the above high field negative magnetoresistance in bulk ferromagnets we have calculated the high field values of $\alpha=1/\rho(\partial\rho/\partial B)$ by fitting the transverse MR vs B data (Fig. 2) to straight lines for $B\geqslant4$ T when the multilayers are in their ferromagnetic state. The high-field slopes (α) are found to be negative at all temperatures for all the samples implying a negative magnetoresistance. Figure 8 shows the α vs T plot for sample 3 along with the error bars. At 300 K the value of $\alpha=-3\times10^{-4}T^{-1}$ is comparable to $-4\times10^{-4}T^{-1}$ for bulk crystalline Fe.[1] The magnitude of the negative slope (α) is found to increase with temperature as expected and has a small value at 4.2 K since there are not too many magnons at such low temperatures that could be quenched by the external magnetic field.

IV. CONCLUSIONS

To summarize, high-field Hall effect data in three Fe-Cr multilayer samples, prepared by Xe-ion beam sputtering, show that the side-jump mechanism is mainly responsible for the R_S. The usual scaling law $R_S=b\rho^2$ for nonclassical transport holds provided ρ is taken as the resistivity in $B=3$ T where the Fe layers are aligned ferromagnetically and not at $B=0$ where the coupling between the Fe layers are antiferromagnetic. The value of b is of the same order as in Fe-Cr dilute alloys. High-field transverse magnetoresistance vs B data, when fitted to a straight line, show that the magnitude of the negative slope increases with temperature as in crystalline Fe due to less electron-magnon scattering at high temperatures.

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