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Anomalous Hall effect in $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$: Universal scaling law and electron localization below the Verwey transition

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We show that the well-established universal scaling $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}^{1.6}$ between anomalous Hall and longitudinal conductivities in the low conductivity regime ($\sigma_{xx} < 10^4 \Omega^{-1} \text{cm}^{-1}$) transforms into the scaling $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}^2$ at the onset of strong electron localization. The crossover between the two relations is observed in magnetite-derived $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films where an insulating/hopping regime follows a bad metal/hopping regime below the Verwey transition temperature T_v . Our results demonstrate that electron localization effects come into play in the anomalous Hall effect (AHE) modifying significantly the scaling exponent. In addition, the thermal evolution of the anomalous Hall resistivity suggests the existence of spin polarons whose size would decrease below T_v . © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4961144>]

Magnetite is the first known magnet and is the prototypical oxide compound for magnetic exchange mechanisms. It is a ferrimagnetic material where the antiparallel alignment of magnetic moments results in a spontaneous magnetization associated with a high Curie temperature (~ 900 K). Corollary, it behaves at room temperature as a half-metallic compound with an expected 100 % spin polarization, and undergoes at the referred to Verwey temperature ($T_v = 120$ K)¹ a first-order metal-insulator transition. Great progress has been achieved from both the theoretical^{2,3} and experimental⁴⁻⁷ points of view for a full understanding of these fascinating properties and the development of magnetite-based storage or spintronic⁸⁻¹¹ devices. The zinc-substituted Fe_3O_4 oxide, namely $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$, has been much less investigated although it offers the possibility to tune the magnetic or electrical properties as a function of the Zn content, while maintaining the essential physical specificities of magnetite.¹²⁻¹⁷

The Hall effect in ferromagnetic compounds has by its own been the subject of intense research (for a review see Ref. 18). For a broad range of materials with magnetization M_z under an applied perpendicular field H_z , the in-plane Hall resistivity ρ_{xy} follows the semi-empirical law:

$$\rho_{xy} = R_O \mu_0 H_z + R_s \mu_0 M_z,$$

where the first term is the ordinary Hall effect (OHE) due to the Lorenz force on charge carriers and the second term is the anomalous Hall effect (AHE). By involving spin-polarized carriers which, unlike magnetic properties, may be controlled electrically, the AHE is essential in spintronics.¹⁹

Three different regimes for the behavior of the AHE as a function of the longitudinal conductivity σ_{xx} have been identified: (i) a high conductivity regime ($\sigma_{xx} > 10^6 \Omega^{-1} \text{cm}^{-1}$) for which the AHE transverse conductivity σ_{xy}^{AHE} varies linearly with σ_{xx} , (ii) an intermediate metallic regime ($10^4 \Omega^{-1} \text{cm}^{-1} < \sigma_{xx} < 10^6 \Omega^{-1} \text{cm}^{-1}$) for which σ_{xy}^{AHE} is independent of σ_{xx} , and (iii) a bad metal/hopping regime ($\sigma_{xx} < 10^4 \Omega^{-1} \text{cm}^{-1}$) for which σ_{xy}^{AHE} varies as σ_{xx}^n where $1.6 < n < 1.7$. Magnetite as well as $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ belongs to this third class of materials.^{20,21} We report here on anomalous Hall effect measurements on $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films, (001) oriented or polycrystalline,

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for which the intrinsic regime predicted by Onoda *et al.*²² and associated to the universal scaling relation $\sigma_{xy}^{AHE} \sim \sigma_{xx}^{1.6}$ is observed above the Verwey transition, while σ_{xy}^{AHE} scales with σ_{xx}^2 at lower temperatures. The two different scaling behaviors match with the two electron transport regimes above and below the Verwey temperature, namely the Arrhenius-like thermally activated regime and the more localized Mott variable range hopping (VRH) regime, respectively. Last, the interplay between itinerant electrons and local magnetic moments is highlighted through the thermal evolution of ρ_{xy}^{AHE} .

The $Zn_xFe_{3-x}O_4$ films were obtained by pulsed laser deposition (PLD) from stoichiometric targets with x about 0.35 on MgO(001) substrates, according to two procedures. Non-stoichiometric (001)-oriented layers were grown at 500°C under 3×10^{-7} mbar O_2 pressure, and polycrystalline stoichiometric layers were obtained by growth at room temperature, followed by annealing at 450°C. The thickness and chemical composition of the films were determined by random Rutherford backscattering spectrometry (RBS). The 82 nm-thick $Zn_xFe_{3-x}O_{3.8}$ (001) films with $x \sim 0.42$ will be referred to as ZFO (001) and the 145 nm-thick polycrystalline $Zn_xFe_{3-x}O_4$ films with $x \sim 0.39$ will be referred to as ZFO poly. A previous study¹⁷ has shown that despite their very different microstructures, the conductivity and ferromagnetic properties of the two types of films are close together. In particular, a smooth (feeble) Verwey-like transition at ~ 120 K is apparent in the thermal evolution of the resistivity for both of them. The ZFO poly film in which the amount of antiphase boundaries (APBs) is considerably lower than in ZFO(001) shows in addition a decrease of the spontaneous magnetization below 120 K. The transport and magnetic experiments were carried out with a Physical Property Measurement System (PPMS). The magnetic field (up to 9 T) was applied along z direction, perpendicular to the (xy) film plane. The different electrical characteristics, namely longitudinal resistivity ρ_{xx} , transverse resistivity ρ_{xy} , magnetoresistance (MR), were obtained from Hall crosses. The films were patterned using optical lithography and wet chemical etching into $180 \times 1560 \mu^2$ Hall bars with a geometry of two $50 \mu\text{m}$ -wide transverse branches (with Au/Cr contact-pads).

The transverse Hall resistivity ρ_{xy} as a function of magnetic field μ_0H is displayed in Fig. 1 for different temperatures. The presence of the two OHE and AHE components is evident in the two types of films. In the polycrystalline case, after the rapid increase of ρ_{xy} in the low field region, one observes a linear dependence as soon as $\mu_0H > 2.5$ T, reflecting the easy complete saturation of magnetization in that type of films. It is more difficult to reach saturation in the ZFO(001) film as a consequence of the presence of APBs, and the (literature admitted) antiferromagnetic coupling between spins at their neighborhood. Importantly, the slope of the high field linear OHE part does not change while temperature decreases from 300 K up to at least 140 K. From this linear high field dependence of $\rho_{xy}(H)$, we may confidently derive the ordinary Hall coefficient $R_O = 1/(n e)$, where n is the electron density. We obtain $n = 5.7 \times 10^{21} \text{ cm}^{-3}$ for the ZFO poly film, *i.e.* 0.42 electron per $Zn_{0.39}Fe_{2.61}O_4$ formula unit (f.u.). Labeling by A and B the tetrahedral and octahedral sites in the spinel structure, respectively, the zinc ferrite f.u. is $(Zn^{2+}_xFe^{3+}_{1-x})_A(Fe^{3+}_{1+x}Fe^{2+}_{1-x})_BO_4^{2-}$. Due to the super-exchange antiferromagnetic coupling between A and B sites and the double-exchange ferromagnetic coupling between cations in B sites, only one minority spin electron from each ($3d^6$) Fe^{2+} cation is involved in the charge transport process [*i.e.* (1- x) spin down electron per f.u.]. At temperatures $> T_v$, the electronic conductivity is due to the delocalized hopping between Fe^{2+} and Fe^{3+} cations in B sites. The lattice parameter in ZFO being $a = 8.409 \text{ \AA}$ and one f.u. occupying the volume $(a/2)^3$, the n value deduced from the Hall $\rho_{xy}(H)$ slope gives 0.42 electron per f.u.. This value very well agrees with the number of Fe^{2+} cations per f.u. as determined by RBS in the ZFO poly film, *i.e.* 0.61. In ZFO(001), we derive 0.23 electron versus 0.58 Fe^{2+} per f.u..

The measurement of the longitudinal resistivity ρ_{xx} as a function of temperature T (curves not shown) gives access to the electron mobility μ from $\rho_{xx} = 1/(n e \mu)$. For temperatures > 120 K, the number n of mobile electrons is constant and is determined by the Fe^{2+} content (see above). At 300 K, we derive μ equal to 0.06 (0.02) $\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in the ZFO(001) (poly) film. The thermal evolutions of the mobility are shown in Fig. 2. An $\exp(-E_a/kT)$ dependence is observed, where E_a is the activation energy for itinerant electron hopping between Fe^{2+} and Fe^{3+} in equally occupied B sites. We find $E_a = 51$ (59) meV for the (001) (poly) film, in accordance with other resistivity measurements on full ZFO layers.¹⁷ In pure magnetite Fe_3O_4 crystals, a charge ordering takes place

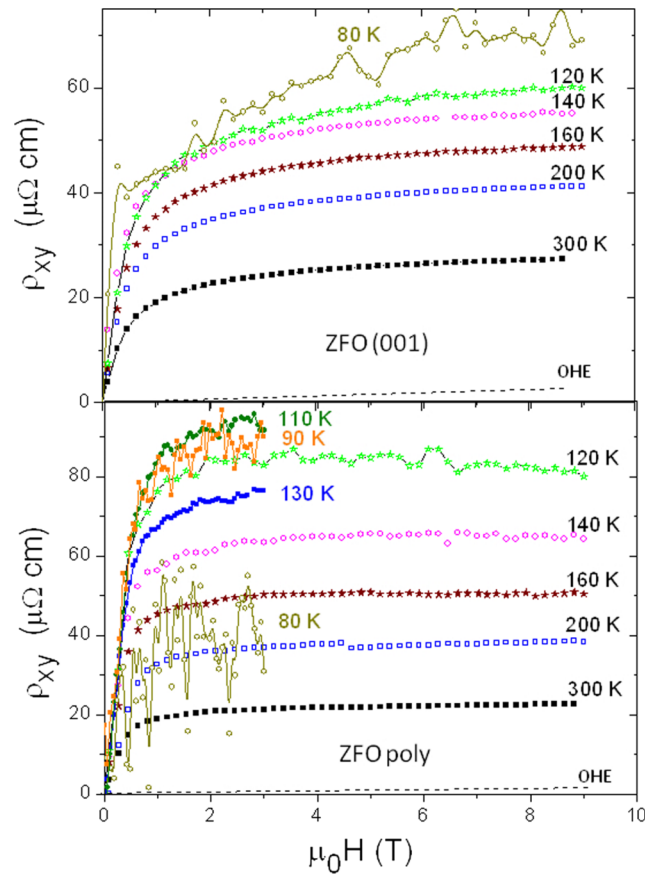


FIG. 1. The transverse resistivity as a function of magnetic field for different temperatures in (001)-oriented and polycrystalline ZFO layers.

below $T_v = 120$ K together with a structure symmetry lowering,^{1,2,6,7} leading to an abrupt increase of the resistivity. Although details on this charge ordering are still a matter of debate, one generally admits that instead of $\text{Fe}^{2.5+}$ mixed-valence undifferentiated ions, the Fe^{2+} and Fe^{3+} ions at $T < T_v$ are localized in specific B sites. In thin films, the transition is either smooth or absent. In

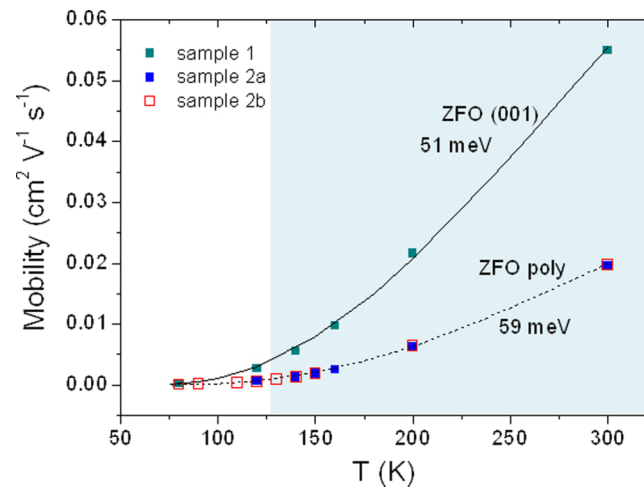


FIG. 2. The electron mobility derived from ρ_{xx} under assumption of a constant density of electrons and fitted according to the Arrhenius model.

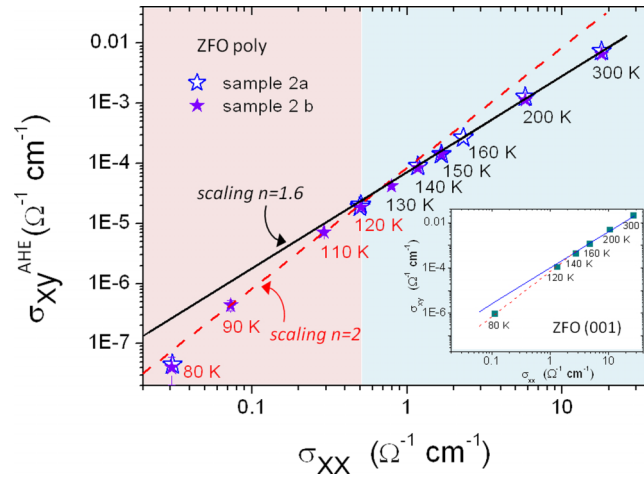


FIG. 3. The anomalous Hall conductivity σ_{xy}^{AHE} of the ZFO poly film as a function of longitudinal conductivity σ_{xx} in log-log scale. The straight (dotted) line features the $\sigma_{xx}^{1.6}$ (σ_{xx}^2) dependence of σ_{xy}^{AHE} . The inset refers to the ZFO(001) film.

our $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ films, a Mott VRH $\exp(T_0/T)^{1/4}$ dependence of the longitudinal resistivity has been clearly evidenced for $80 \text{ K} < T < 120 \text{ K}$.^{17,23} This reverts to consider in $\sigma_{xx} = 1/\rho_{xx}$ a tunnel-type hopping term between distant localized sites and n and μ cannot be considered independently.

The AHE contribution to transverse resistivity, simply referred to in the following as ρ_{xy}^{AHE} is generally deduced from $\rho_{xy}(H)$ at high fields, where magnetization saturates, removing the ordinary linear part. In our case, one may note that the OHE contribution is very small with respect to the AHE part, making safely the determination of $\rho_{xy}^{\text{AHE}} = R_s \mu_0 M_s$, where M_s is the saturation magnetization. From ρ_{xy}^{AHE} one may derive the AHE transverse conductivity σ_{xy}^{AHE} , often called the anomalous Hall conductivity, accordingly to $\sigma_{xy} = \rho_{xy}/(\rho_{xx}^2 + \rho_{xy}^2)$. Fig. 3 displays the magnitude of σ_{xy}^{AHE} as a function of σ_{xx} in double logarithmic scale for two different crosses (samples 2a and 2b) from the ZFO poly film. The error bars on σ_{xy}^{AHE} are lower than the width of the symbols used in the representation, except for one point at 80 K. For temperatures $T > 120 \text{ K}$, the Hall conductivity scales nicely with the longitudinal conductivity accordingly to the relationship $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}^{1.6}$. For $T \leq 120 \text{ K}$, the data deviate obviously from the scaling with exponent 1.6 and the relationship appears as $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}^2$. Note the point at 80 K is slightly lower than expected from σ_{xx}^2 , suggesting a slightly higher scaling exponent. No measurement could be performed at $T < 80 \text{ K}$ due to the huge values of resistance ($> 6 \times 10^6 \Omega$) reached in this highly insulating range.

From the theory developed for multi-band ferromagnetic metals with dilute impurities, in the regime of low conductivity, the scaling relation with exponent 1.6 corresponds to the intrinsic regime with follows the extrinsic (skew scattering) regime where $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}$.²² Similarly, an average exponent 1.65 was calculated for the Hall effect in quantum Hall insulators.²⁴ Experimentally, the scaling with exponent 1.6 has been reported, over five decades of the longitudinal conductivity (from 10^{-2} up to $10^4 \Omega^{-1} \text{ cm}^{-1}$), for very different materials including metallic Fe ultra-thin films²⁵ and compounds in which the conduction regime may being varied from metallic to hopping: (La,Sr)MnO₃, (Ti,Co)O₂, (Ga,Mn)As.^{26–29} The scaling $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}^n$ with $n = 1.6$ has also been found in pure magnetite films²⁰ while a $n = 1.69$ exponent has been derived for zinc ferrites films and Fe-ZnO granular ones.^{21,30} These converging experimental findings on so various materials give strong support to the universal character of the 1.6 scaling relation, even if a complete AHE theory including the insulating regime is still missing.

Thanks to the large variation in its longitudinal conductivity σ_{xx} by the sole mean of temperature, the $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ film system is well adapted to identify AHE differences between the bad metal/thermally activated regime ($T > T_v$) and the more insulating/localized VRH regime ($T < T_v$). The high crystalline quality of our ZFO poly film allows highlighting the crossover between these two regimes in the anomalous Hall conductivity at about the Verwey temperature ($\sim 120 \text{ K}$). The scaling relation clearly transforms from $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}^{1.6}$ to $\sim \sigma_{xx}^2$. The ZFO (001) film in which the

Verwey transition was not detected in the thermal dependence of magnetization¹⁷ shows the same trend of a significant increase of the exponent at low temperatures (see inset of Fig. 3). A change from $n = 1.6$ to 2 has recently been observed at very low temperature in ultra-thin ferromagnetic “metallic” CNI_3 films, where strong two-dimensional localization effects intervene when reducing the film thickness.³¹ A similar study on ultra-thin FePt films³² has put forward that disorder-induced electron localization instead of Coulomb interaction is responsible for this $n = 2$ scaling. Hall data on anatase $\text{Ti}_{1-x}\text{Co}_x\text{O}_2$ films³³ also indicated a significant increase of the scaling exponent in the low T range, although this effect had not been discussed by the authors. The change of scaling n across an electronic transition driven by electron localization effects, whether they are driven by the temperature or by the thickness, leads us to conclude that universal quantum corrections are in play, which in the context of spin-dependent scattering processes still remain to be elucidated.

In Fig. 4(a), we display the $\rho_{xy}(H)$ and $M(H)$ curves of the ZFO poly sample at 300 and 150 K. The saturation magnetization at 300 K (493 kAm^{-1}) corresponds to $3.95 \mu_B$ per f.u., that is 62 % of the $(4+6x)$ value expected in ideally ordered $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$. The proportionality between ρ_{xy} and M is confirmed and the hysteresis features at 150 K are retrieved in the Hall measurements. However, whereas the saturation magnetization M_s increases by a factor 1.1 between 300 and 150 K, accordingly with the Bloch’s law,¹⁷ the Hall resistivity increases by a factor 2.6. Consequently, the Hall resistivity ρ_{xy}^{AHE} is sensitive to thermal effects that are not detected by the macroscopic M measurement. It is thus interesting to study in details the $\rho_{xy}^{\text{AHE}}(T)$ dependence more especially as the ρ_{xy}^{AHE} data are directly accessible by the experiment whereas the $\sigma_{xy}^{\text{AHE}}(T)$ values are derived from $\sigma_{xy} = \rho_{xy}/(\rho_{xx}^2 + \rho_{xy}^2) \sim \rho_{xy}/\rho_{xx}^2$ with (i) an over-exceeding contribution and (ii) a steeper

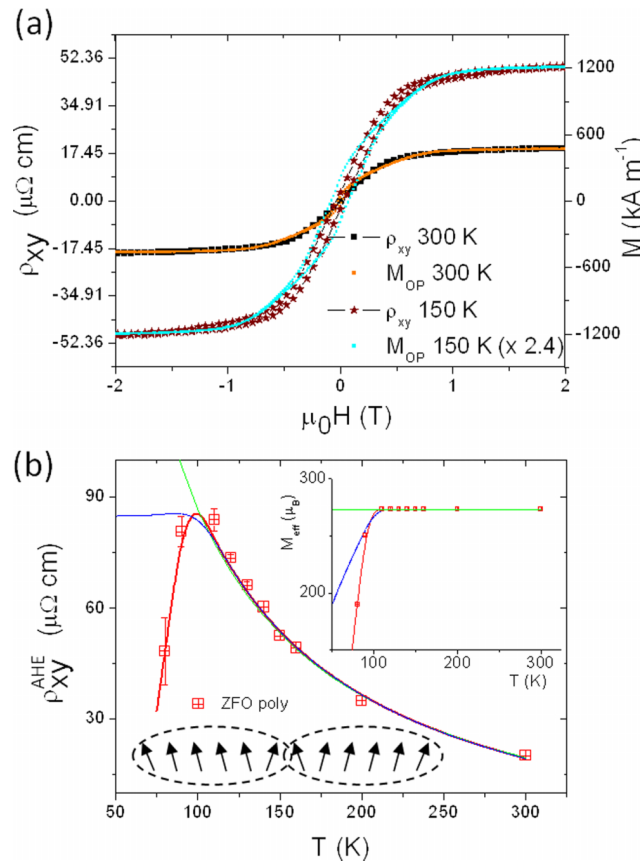


FIG. 4. (a) Comparison between $\rho_{xy}(H)$ (left scale) and $M(H)$ (right scale) measurements at 300 and 150 K. (b) The anomalous Hall resistivity (symbols) at different temperatures with respect to simulation (solid lines) using $\rho_{xy}^{\text{AHE}}(T) = m_{\text{eff}} \mathcal{Q}$ where m_{eff} is as indicated in the inset. An illustration of two spin polarons inside which the parallelism of magnetic moments of Fe^{2+} cations induce delocalization of Fe^{2+} minority spin electrons (magnetic moments from Fe^{3+} cations are not represented).

dependence on T of ρ_{xx} with respect to ρ_{xy} . The ZFO poly $\rho_{xy}^{\text{AHE}}(T)$ values are displayed in Fig. 4(b). In the range $\sigma_{xx} > 0.1 \Omega^{-1} \text{cm}^{-1}$ ($T \geq 110 \text{ K}$), they follow a $1/T$ dependence, reminiscent of the Curie-type susceptibility of a paramagnetic system. In the range $\sigma_{xx} \leq 0.1 \Omega^{-1} \text{cm}^{-1}$ ($T < 110 \text{ K}$), the ρ_{xy}^{AHE} values hardly increase anymore and even decrease. We found a similar behavior in ZFO(001) (data not shown). Saturation of ρ_{xy}^{AHE} has been observed in the strongly localized regime of ultra-thin “metallic” films^{31,32} while a decrease of ρ_{xy}^{AHE} below 110 K has been reported in the case of the $\text{Ti}_{1-x}\text{Co}_x\text{O}_2$ system.³³ In the following, we suggest some possible explanations for the overall T -dependence of ρ_{xy}^{AHE} .

For $T > T_v$, the spin-down itinerant electrons mediate the double-exchange ferromagnetic coupling between magnetic moments in B sites; electron hopping is thus intimately related to the local magnetization state, especially at low field. We refer to the “spin polaron” picture^{34,35} to describe our ZFO system. The model has already been used with success to fit the MR data.¹⁷ It reverts to consider “giant molecules” bearing giant magnetic moments m_{eff} , associated to ideal regions where nearest neighbors Fe^{2+} ions have their moments perfectly aligned and where itinerant hopping is thus promoted. The different “spin polarons” align themselves parallel only at high field, and at high T , electrons may tunnel between them. At low field, the m_{eff} moments of the “spin polarons” are randomly oriented. We have here directly compared $\rho_{xy}^{\text{AHE}}(T)$ to $[m_{\text{eff}} \mathfrak{L}(m_{\text{eff}} \mu_0 H/k_B T)]$ where \mathfrak{L} is the Langevin function, considering the low field limit ($H \sim 0$). As shown in Fig. 4(b), we nicely fit the ZFO poly $\rho_{xy}^{\text{AHE}}(T)$ data in the 120-300 K range by means of a single parameter m_{eff} equal to $273 \mu_B$, supporting the “spin polaron” model. For ZFO(001), the m_{eff} parameter has been found equal to $100 \mu_B$. The average giant magnetic moment is of the order of $200 \mu_B$ and corresponds to a “giant molecule” of 50 Fe^{2+} ions, whose width is about 1.6 nm. This gives $\sim 3.7 \text{ Fe}^{2+}$ ions ferromagnetically coupled along one direction, each ion bearing $4 \mu_B$. This “spin polaron” size (1.6 nm) is of the same order than the average distance between Zn^{2+} ions in the ZFO films ($\sim 1.1 \text{ nm}$) as it may be deduced from the RBS chemical composition. The “spin polaron” width may be viewed as the localization length ξ the electron wave packets, each Fe^{2+} ion providing one electron. Apart from grain boundaries, the Zn^{2+} ions are the main source of weakening of the magnetic order and they provide here the upper limit to the wave packet extension.

With decreasing T ($< T_v$), the electrons become more and more localized (trapped) inside a “spin polaron”. In other words, the average size of a “spin polaron” is expected to shrink since more and more magnetic moments may be misaligned due to electron localization. It has recently been demonstrated in magnetite that, below T_v , the localization occurs within linear three-Fe-site units, called “trimerons”, or within interconnected “trimerons”.^{7,36} A “trimeron” involves only one Fe^{2+} and it may be viewed as the smallest form of a “spin polaron”. By assuming from 110 K a decreasing m_{eff} with decreasing T (see inset in Fig. 4(b)), the $[m_{\text{eff}} \mathfrak{L}(m_{\text{eff}} \mu_0 H/k_B T)]$ function allows reproducing the saturation as well as the decrease of $\rho_{xy}^{\text{AHE}}(T)$, according to the $m_{\text{eff}}(T)$ slope value. While saturation of $\rho_{xy}^{\text{AHE}}(T)$ is expected for a n scaling exponent exactly equal to 2, the decrease of $\rho_{xy}^{\text{AHE}}(T)$ would suggest a slightly higher n value. It is possible that ρ_{xy}^{AHE} involves intrinsic and extrinsic contributions both relevant at low temperatures, as it has been demonstrated in the case of ultra thin metallic films.³⁷

We confirm the universal character of the $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}^{1.6}$ scaling relation between anomalous Hall and longitudinal conductivities in the bad metal/hopping electron transport regime. However, below the Verwey temperature which characterizes the metal/insulator transition in $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ films, the anomalous Hall data clearly deviate from the 1.6 exponent scaling. By moving from a partly delocalized metallic regime towards a localized insulating one, the scaling relation transforms into $\sigma_{xy}^{\text{AHE}} \sim \sigma_{xx}^2$. This finding could motivate further theoretical studies on AHE in low conductivity ferromagnetic materials, and more generally in strongly correlated electron systems that are important for spintronics development. Finally, we show that the anomalous resistivity restitutes spin-dependent electron localization effects.

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