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Universal scaling of the anomalous Hall effect in Fe_3O_4 epitaxial thin films

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We report a systematic study of the anomalous Hall effect in epitaxial thin films of magnetite grown on MgO (001) substrates as a function of film thickness (from 5 to 150 nm) and temperature ($60\text{ K} < T < 300\text{ K}$). The Hall resistivity was found to be negative for all the measured temperature range, presenting a huge enhancement at temperatures below the Verwey transition, reaching values above 1 mΩ cm. The anomalous Hall conductivity scales as $\sigma_H \propto \sigma_{xx}^{1.6}$ over four decades of the longitudinal conductivity σ_{xx} in all the samples, in agreement with a recent unified theory for the anomalous Hall effect.

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In *magnetic* materials the Hall resistivity ρ_H is given by $\rho_H = R_O\mu_0 H + R_A\mu_0 M$. The first term, proportional to the applied magnetic field, describes the ordinary Hall effect (OHE) and the second, in general much larger than the first one, is the anomalous Hall effect (AHE), which is proportional to the magnetization of the material. The OHE is caused by the Lorentz force acting on moving charged carriers, whereas the origin of the AHE has been a controversial issue for decades, where theories based on intrinsic[1,2] or extrinsic contributions[3,4] tried to explain this effect. Different dependences between the Hall and the longitudinal resistivity ρ_{xx} are given, depending on which origin is associated. If an intrinsic mechanism, based on the Berry phase of Bloch waves, dominates the effect, a dependence $\rho_H \propto \rho_{xx}^2$ should be followed, whereas for the skew scattering or the side jump, $\rho_H \propto \rho_{xx}$ or $\rho_H \propto \rho_{xx}^2$ are expected, respectively. A recent theory[5] based on multiband ferromagnetic metals with dilute impurities seems to have solved this complex scenario, where three regimes can be distinguished as a function of the longitudinal conductivity. In the clean regime with extremely high conductivity the skew scattering causes the effect ($\sigma_H \propto \sigma_{xx}$). An extrinsic-to-intrinsic crossover occurs at lower conductivities ($\sigma_{xx} = 10^4 - 10^6\ \Omega^{-1}\text{ cm}^{-1}$), where σ_H becomes constant. In the dirty regime ($\sigma_{xx} < 10^4\ \Omega^{-1}\text{ cm}^{-1}$) a relation $\sigma_H \propto \sigma_{xx}^{1.6}$ is predicted, caused by the damping of the intrinsic contribution. Recently, this crossover has been experimentally found for a series of itinerant ferromagnets[6] and a compilation of an appreciable amount of low-conductivity compounds reveal the expected dependence in the dirty limit, regardless of hopping or metallic conduction.[7] However, a theory explaining such behavior for materials dominated by hopping conduction is still not available.

In this article, we report on a systematic study of the AHE in epitaxial Fe_3O_4 thin films within a wide range of thicknesses (5–150 nm) and in the temperature range from 300 K down to 60 K. The longitudinal conductivity varies over four orders of magnitude ($10^{-2}\ \Omega^{-1}\text{ cm}^{-1} < \sigma_{xx} < 10^2\ \Omega^{-1}\text{ cm}^{-1}$). In all cases the Hall conductivity approximately scales with the longitudinal conductivity as $\sigma_H \propto \sigma_{xx}^{1.6}$, as is expected in the dirty regime. Comparison of our results with others in literature for magnetite demonstrates the universality of this dependence for this material, irrespective of the different mechanisms responsible for the electronic transport, density of defects, stress, or preparation method.

Fe_3O_4 thin films were grown on MgO (001) substrates by pulsed laser deposition in an ultrahigh vacuum system with a background pressure below 5×10^{-9} Torr. Structural characterization including x-ray diffraction, including electron diffraction, and transmission electron microscopy imaging reveal a 001-oriented epitaxial growth as well as a high crystallinity of the films.[8,9] X-ray reflectivity was used for the measurement of the film thickness within a ±0.2% estimated accuracy. Reciprocal space maps measured around the 226-Fe_3O_4 and 113-MgO reflections show a fully strained growth up to at least 150 nm thickness, with no relaxation of in-plane epitaxial stress.[9] Magnetic measurements were made in a superconducting quantum interference device magnetometer, giving saturation magnetization values about 5–15% smaller than in bulk samples, as is usual in similar high quality thin films.[10]

For electrical transport measurements a two-step lithography process was carried out so that a well-defined geometry minimized offset voltages in the measurements. Offset voltages can fully spoil the Hall measurements in systems with large longitudinal resistance (as occurs in the studied films at low temperature), so special care was taken to minimize them. The typical electrode for the flow of the current was 300 μm wide and pads were patterned for the measurements of the voltage drop. For further details, see Ref. 8. These measurements were made using a 6220 dc current source-2182A nanovoltmeter combined Keithley system, using the available delta mode to eliminate the possible thermal potentials from the measurements, as well as sample heating. A closed-cycle refrigerator that allows varying the temperature from 300 to 10 K was used, together with an electromagnet delivering a magnetic field up to 11 kOe. In the present study, we report measurements at room temperature for films with thickness of 150, 40, 20, 9, and 5 nm, as well as a function of the temperature in the 40- and 20-nm-thick films. The values for the resistivity at room temperature measured by the four-probe method increase monotonically for decreasing film thickness. This behavior is mainly associated with the increase of antiphase boundaries (APB) density as the films decrease in thickness.[11,12] APBs are structural de-

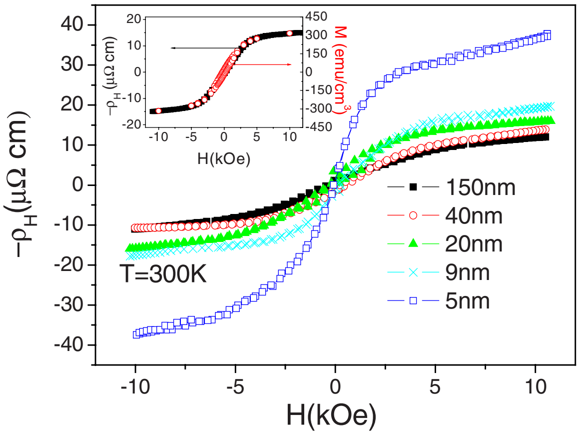

FIG. 1. (Color online) Hall resistivity at room temperature as a function of thickness for a series of samples. The field is applied perpendicular to the plane of the film. The inset shows a comparison of the Hall resistivity at 300 K for a 40-nm-thick film together with the perpendicular magnetization at that temperature for another sample of the same thickness grown in the same conditions. The perfect proportionality between both shows that the AHE dominates the measurement in the range of field measured.

fects, formed during the growth process, which are unavoidably present when growing $\mathrm{Fe_3O_4}$ epitaxial thin films.[11,12] Typical resistivity values ranging from 5.46 mΩ cm for the 150 nm film to 121.4 mΩ cm for the 5 nm film have been obtained. These values are comparable to other studies,[10–13] confirming the high quality of the films.

In Fig. 1 we represent the Hall resistivity ρ_H at room temperature as a function of the film thickness. For all samples, ρ_H is negative, increasing in modulus as the films get thinner. The value for the 40-nm-thick film at 11 kOe is 13.9 $\mu\Omega$ cm, in excellent agreement with that obtained in Ref. 10 at the same field (13.75 $\mu\Omega$ cm), as expected for similar high-quality films. The maximum Hall slope at zero field is found for the 5-nm-thick film, reaching 125 $\mu\Omega$ cm/T, which corresponds to a sensitivity of 250 V/AT. This sensitivity at room temperature is only higher in ferromagnetic materials for $\mathrm{Co}_x\mathrm{Fe}_{1-x}$/Pt multilayers.[14] It is important to remark that in the range of magnetic fields measured, the contribution of the AHE dominates completely the Hall effect, being unrealistic to determine the ordinary part from the Hall slope at the highest measured magnetic field. This impossibility to saturate the film in moderate magnetic fields is due to the antiferromagnetic coupling between spins at the neighborhood of APBs.[15] This fact can explain the dispersion in values for the ordinary Hall constant R_O obtained by different authors,[10,16] as was pointed out before.[8] Therefore, there should exist a perfect proportionality between ρ_H and the magnetization perpendicular to the plane of the film. This is clearly seen, e.g., for the 40-nm-thick film at room temperature in the inset of Fig. 1. According to our room-temperature Hall effect measurements in the 40 nm film up to 90 kOe (not shown here), we estimate that the ordinary Hall contribution to the total Hall resistivity under 10 kOe is below 0.15 $\mu\Omega$ cm. Thus, we are safe to conclude that the AHE dominates the observed Hall effect and we can neglect any relevant influence of the OHE in the present results. Such measurements also indicate that even under 90 kOe the anomalous Hall effect is not fully saturated as was previously observed in magnetization measurements under high fields.[15]

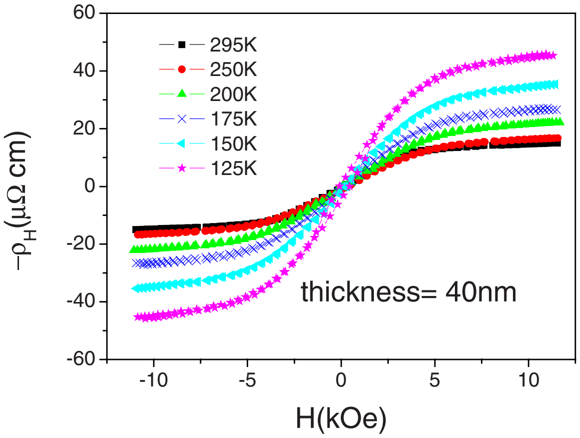

FIG. 2. (Color online) Hall resistivity isotherms in a film of 40 nm for temperatures above the transition. The Hall resistivity increases as the temperature is diminished, reaching in this case 45 $\mu\Omega$ cm at 125 K.

In order to account for the thermal dependence of ρ_H we have measured the 20 and 40 nm films as a function of the temperature. The temperature where the Verwey transition occurs is associated with a jump in the longitudinal resistivity curve by a factor of about 100 at $T_\mathrm{v} \sim 120$ K for bulk samples. In the case of thin films, the transition is much less abrupt and takes place at slightly lower temperatures, which is generally explained by the stress created by the substrate[17] or oxygen nonstochiometry.[18] The transitions occur at 110 and 108 K for the 40- and 20-nm-thick films, respectively. These temperatures are the same as in magnetic measurements where the change in the slope is more abrupt. In Fig. 2 we show the ρ_H isotherms for the 40-nm-thick film at temperatures above the Verwey transition. For both samples ρ_H increases in modulus monotonously when the temperature diminishes, presenting a huge enhancement as the temperature approaches the transition. This abrupt change in ρ_H at T_V is a general behavior for $\mathrm{Fe_3O_4}$, which seems to be related with changes in the electronic structure and the spin-orbit coupling that occurs at the transition.[18] In both samples the values for ρ_H increase by a factor higher than 100 from room temperature down to 60 K, reaching values above 1 mΩ cm at that temperature.

In Fig. 3 we show the absolute value of the Hall conductivity at maximum field (11 kOe) as a function of the longitudinal conductivity for all the samples measured. In spite of the different thicknesses and measurement temperature, the relation $\sigma_H \approx 10^{-4}\sigma_{xx}^{1.6}$ (dashed line) is followed in all cases. The increase in the value of the error bars for σ_H at temperatures below the transition ($\sigma_{xx} < 1.5\ \Omega^{-1}\ \mathrm{cm}^{-1}$) is due to the fact that residual contributions to the voltage measured, such as the longitudinal resistivity, increase substantially. Another relevant source of errors due to slight unavoidable misalignments in the experimental setup is the contribution of the planar Hall effect, which has recently been found to be giant

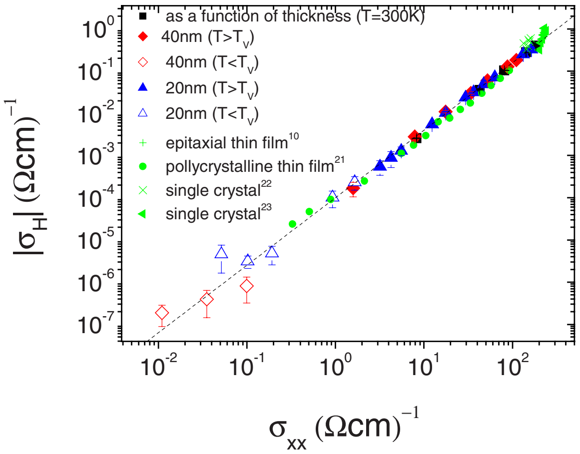

FIG. 3. (Color online) Relationship between the magnitude of the Hall conductivity $[|\sigma_H| = \rho_H/(\rho_{xx}^2+\rho_H^2)]$ at 11 kOe and the longitudinal conductivity $[\sigma_{xx}=\rho_{xx}/(\rho_{xx}^2+\rho_H^2)]$ for our films and data taken from Refs. 10 and 21–23. Dashed line is the function $f(\sigma_H)=10^{-4}\,\sigma_{xx}^{1.6}$.

in epitaxial films of Fe_3O_4,[19] and could be at the origin of previous results yielding a different sign of ρ_H (Ref. 20) in this temperature range. Together with our results, other measurements found in the literature for the AHE in magnetite have also been plotted. Specifically, another 45-nm-thick epitaxial thin film at room temperature,[10] a polycrystalline 250-nm-thick film at temperatures below 300 K (Ref. 21), and single crystals[22,23] in the range $150\text{ K} < T < 500\text{ K}$ have been included. These data approximately converge with ours, revealing the same physical origin in all cases.

The fact that Fe_3O_4 has been predicted to be half metal,[24] with a large Curie temperature (860 K),[18] makes it a suitable material for application in spintronics. Therefore, much effort has been done by many groups to grow and characterize high-quality epitaxial thin films, revealing substantial differences between some properties of the bulk material and the films, such as the magnetization, electrical resistivity, or the magnetoresistance ratios.[12,15–17] These differences have been associated to the films microstructure and the presence of antiphase boundaries, becoming more evident the thinner the films are, because the density of APBs varies as $t^{-1/2}$, where t is the thickness of the film.[11,12]

In this paper we show general results for the AHE in magnetite. The enhancement in the Hall resistivity as the temperature is diminished seems to be related with the increase in the longitudinal resistance, as a consequence of the broadening that occurs in the band gap as T_V is approached. This results in huge Hall resistivities of the order of mΩ cm at low temperatures. The dependence $\sigma_H \propto \sigma_{xx}^{1.6}$ found over four decades of longitudinal conductivity is in accordance with a recent unifying theory for the AHE.[5] This relation is followed by samples in a wide range of thicknesses and irrespective of the measurement temperature. Furthermore, the same dependence for σ_H is found in other samples of magnetite taken from the literature, regardless if they are single crystals or polycrystalline thin films. The theory[5] explains that the magnitude of the AHE is determined by the degree of resonance caused by the location of Fermi level around an anticrossing of band dispersions. Besides, it assumes metallic conduction. From our results we can infer that the Verwey transition, associated with a cubic-to-monoclinic structure transition, does not play an important role in this behavior. It was pointed out before that this dependence is also followed experimentally in hopping conduction,[7] consistently with calculations of the quantum Hall effect in disordered systems with hopping conduction.[25] In the case of Fe_3O_4, our results suggest that this dependence also takes place irrespective of the different conduction mechanisms occurring above and below T_V.[18] Another important issue is related with the presence of APB. As was mentioned before, some magnitudes such as the resistivity or magnetization are subjected to important changes from the bulk material to the thin films, due to the presence of these structural defects. The moderate magnetoresistance ratios in films are explained by the spin-polarized transport through them. The AHE is affected by the density of APBs through the change in the longitudinal conductance, which decreases as the film thickness does. Finally, it should also be remarked that the presence of stress in epitaxial thin films caused by the substrate could eventually have an influence in the band structure in comparison with the bulk material, but the convergence of all data reveals that this is not an important issue for the AHE. All these facts support the belief that the relation $\sigma_H \propto \sigma_{xx}^{1.6}$ is universal for this low-conductivity regime.

In summary, we report a general study of the anomalous Hall effect in high-quality epitaxial thin films of magnetite. In over four decades of longitudinal conductivity we find the dependence $\sigma_H \propto \sigma_{xx}^{1.6}$, irrespective of thickness or measurement temperature. Comparison with literature confirms this behavior is general for this compound. This result supports a recent theory developed to explain the anomalous Hall effect, indicating the universality of this relation in the dirty regime of conductivities.

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