AIP Applied Physics Letters

Transversal magneto-resistance in epitaxial Fe3O4 and Fe3O4/NiO exchange biased system

Han-Chun Wu, R. Ramos, R. G. S. Sofin, Zhi-Min Liao, M. Abid et al.

Citation: Appl. Phys. Lett. **101**, 052402 (2012); doi: 10.1063/1.4739951
View online: http://dx.doi.org/10.1063/1.4739951
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v101/i5
Published by the AIP Publishing LLC.

APPLIED PHYSICS LETTERS **101**, 052402 (2012)

Transversal magneto-resistance in epitaxial Fe_3O_4 and Fe_3O_4/NiO exchange biased system

Han-Chun Wu,[1,a)] R. Ramos,[1] R. G. S. Sofin,[1] Zhi-Min Liao,[2] M. Abid,[3] and I. V. Shvets[1]
[1]*CRANN and School of Physics, Trinity College Dublin, Dublin 2, Ireland*
[2]*State Key Laboratory for Mesoscopic Physics, Department of Physics, Peking University, Beijing 100871, People's Republic of China*
[3]*Ecole Polytechnique Federale de Lausanne/IPMC/LPMN, Station 3, CH 1015, Switzerland*

(Received 23 February 2012; accepted 16 July 2012; published online 30 July 2012)

We have investigated transversal magneto-resistance (MR) in epitaxial Fe_3O_4 and Fe_3O_4/NiO exchange biased systems. It was found that the magnetic field dependence and the magnitude of the transversal MR in both systems strongly depend on the bias current density which suggests that the transversal MR in metal oxide with anti-phase boundaries (APBs) cannot be described by the conventional transversal MR for a single magnetic domain. The effect of electron scattering at the APBs may have to be considered. Angular dependence of the transversal MR at low temperature further indicates that the current explanation of the origin of transversal MR on the basis of anisotropic MR alone may not be sufficient for a system experiencing charge ordering. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4739951]

Spintronics focuses on the study of the effects of the spin degree of freedom on the electron transport. These effects have major implications from a fundamental point of view[1] as well as a vast range of possible applications spanning from magnetic recording read-heads[2] to micro compasses[3] and bio-detection devices.[4] Common magnetoresistance (MR) sensors employ the giant magnetoresistance (GMR) or anisotropic magnetoresistance (AMR) effects. The sensitivity of the AMR signal is limited by thermal fluctuations and drift.[5,6] The drawbacks associated with longitudinal AMR measurements can be greatly overcome by measuring the voltage change in the transversal direction instead.[7–9] The origin of the transversal MR is believed to be closely related to the AMR,[7] which is due to anisotropic electron scattering induced by the spin-orbit interaction.[10] In a simple but common situation for a ferromagnetic (FM) film with a single in-plane magnetic domain, the angular dependencies of the longitudinal and transversal resistivities are given by

$$\rho_{xx} = E_{xx}/j = \rho_\perp + (\rho_\parallel - \rho_\perp)\cos^2\theta, \tag{1}$$

$$\rho_{xy} = E_{xy}/j = (\rho_\parallel - \rho_\perp)\sin\theta\cos\theta, \tag{2}$$

where θ is the angle between the applied electric current ($\boldsymbol{j}$) and the magnetization ($\boldsymbol{M}$) and $\rho_\parallel$ ($\rho_\perp$) are the resistivities parallel (perpendicular) to the in-plane magnetization. Equations (1) and (2) represent the AMR and conventional transversal MR (or planar Hall Effect), respectively. Note, in the conventional transversal MR, the voltage V_{xy} is proportional to the current. However, recent reports show that Eqs. (1) and (2) fail to fully account for the magnetotransport properties of crystalline systems.[8,11–13] The suggested equations in the case of crystalline systems with growth direction along [001] are

$$\rho_{xx} = A\cos(2\alpha - 2\varphi) + B\cos(2\alpha + 2\varphi) + C\cos(4\alpha) + D, \tag{3}$$

$$\rho_{xy} = A\sin(2\alpha - 2\varphi) - B\sin(2\alpha + 2\varphi), \tag{4}$$

where α and φ are the angles between $\boldsymbol{M}$ and $\boldsymbol{j}$ relative to the [100] crystal direction, respectively.[8,12,13] In their analysis, Naftalis *et al.* have shown that the magnetic field and temperature dependence of the four fitting parameters A, B, C, and D suggests the need for a more precise microscopic theory for magnetotransport properties.[8]

The transversal MR has been used as a tool to study in-plane magnetization processes,[14] sensing of low magnetic fields,[5] magnetic micro-bead detection,[15] and compass applications.[3] The possibility of using this effect in non-volatile memory devices[16] has also been considered. Schuhl *et al.* demonstrated the advantage of transversal MR measurements in reducing the temperature drift in MR sensors by at least four orders of magnitude and hence potential for a considerable increase in the resolution.[5] Previous measurements in magnetite thin films have shown a large transversal MR response,[17] with a signal several orders of magnitude higher compared to metallic samples and a sensitivity of 400 Ω/T,[18] compared to values of 340 Ω/T for single layer NiFe sensors.[19] This indicates the potential of Fe_3O_4 films for the realization of transversal MR sensors.

Fe_3O_4 is an archetypal oxide with fascinating electrical and magnetic properties. Its high Curie temperature (858 K) and the expected nearly fully spin polarized electron band at the Fermi level make it a strong candidate for potential spintronics devices although initial efforts in exploiting its half metallic nature in magnetic tunnel junctions (MTJ) have been far from promising.[20–23] Therefore, elucidating the mechanisms which affect the magneto-transport in magnetite is of fundamental importance to understand the below expectation tunneling magnetoresistance (TMR) values. Magneto-transport is also important for understanding the conduction

---

[a)] Author to whom correspondence should be addressed. Electronic mail: wuhc@tcd.ie.

mechanism and its metal to insulator transition (Verwey transition). Epitaxial Fe_3O_4 films are usually grown in MgO templates. This system presents anti-phase boundaries (APB), they are defects naturally arising due to the difference in rotational and translational symmetry between substrate and thin film.[24–28] The APBs dominate the transport properties in thin films due to the predominantly antiferromagnetic (AF) exchange they induce.[27,28]

In this letter, we have investigated the dependence of the transversal MR on the bias current in epitaxial Fe_3O_4 and Fe_3O_4/NiO exchange biased system. It was found that the transversal MR for both systems strongly depends on the bias current. The transversal MR ratio is almost constant at low bias and peaks at a critical bias where the d^2V_{xy}/dI^2 shows a peak. Further increasing the bias increases the change in resistance caused by planar magnetic field but decreases the transversal MR ratio. The effect of electron scattering at the APB is considered. We also investigated the angular dependence of the transversal MR at low temperature and further compared it with the AMR. Our analysis suggests that the transversal MR in magnetite cannot be explained by the conventional transversal MR in the single domain case, and the existing theory based on AMR alone is insufficient to explain transversal MR in magnetite.

Our study was performed on a fully epitaxial Fe_3O_4 and a Fe_3O_4/NiO bilayer structure, and the purpose of including the AF NiO layer is to study the effects of the interfacial exchange interaction[29,30] and APB density on the transversal MR properties. The layers were grown on the same MgO (001) single crystal substrate using a shadow mask inside the molecular beam epitaxy system (MBE). This is to assure that the growth conditions inside the system are the same for single layer and bilayer samples. Details of the growth conditions used are given elsewhere.[23] Reflection high energy electron diffraction (RHEED) was employed to confirm the epitaxial growth and establish the growth mode. Fig. 1 shows the RHEED patterns recorded in [100] azimuth during growth which indicate the epitaxial growth of the NiO and the pseudomorphic growth of Fe_3O_4.[23] A high-resolution x-ray diffractometer was also used to confirm the single phase structural and epitaxial nature of the Fe_3O_4 films.

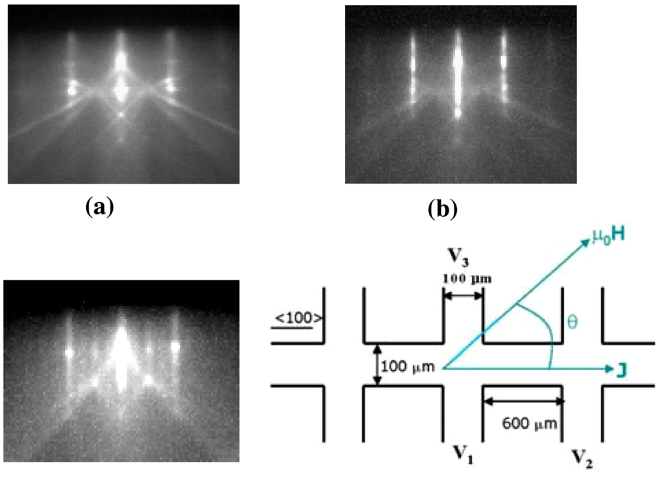

FIG. 1. RHEED images of (a), UHV annealed MgO substrate (b), 15 nm NiO(001) grown on MgO (001) (c), after growth of 10 nm Fe_3O_4(001) on NiO/MgO. The images were recorded in (100) azimuth. (d) Schematic of the Hall bar used for the transversal MR measurements (see Ref. 6).

The transversal MR of the Fe_3O_4 layers and the Fe_3O_4/NiO bilayers was examined using a quantum design physical property measurement system (PPMS). The magnetic field was applied in-plane along varied directions and swept over a loop range between -1 T and $+1$ T. Prior to the transversal MR measurements, the samples were patterned into the Hall bar geometry by UV-lithography and chemically etched.[6] The longitudinal voltage (V_{xx}) and transversal voltage (V_{xy}) were defined by $V_{xx} = V_1 - V_2$ and $V_{xy} = V_3 - V_1$, respectively (see Fig. 1(d)). The current is applied along the long axis of the strips in the [100] direction. The angle between the applied field and the current direction is θ. For the Fe_3O_4/NiO bilayer samples, the exchange biasing field direction is set along the current direction via field cooling process.

Fig. 2 shows the results of the transversal MR measurements performed at room temperature for a 10 nm Fe_3O_4 film (Fig. 2(a)) and 10 nm Fe_3O_4/NiO bilayer (Fig. 2(b)) for different angles between electric current and applied magnetic field with an applied bias current of 10 μA. For comparison, the magnetization reversal process along [100] direction of Fe_3O_4 layer and Fe_3O_4/NiO bilayer at 300 K was also examined using an alternating gradient field magnetometer. One can see from Figs. 2(c) and 2(d) that the transversal MR is an effective method to measure the magnetization reversal process. Moreover, the transversal MR for the Fe_3O_4/NiO bilayer is asymmetric with the applied magnetic field which is different from the case for the single Fe_3O_4 layer. It was well established that the FM layer reversal in FM/AF bilayer system is mainly controlled by FM exchange interaction between the neighboring FM domains and the interfacial interaction due to the AF layer underneath. The interfacial interaction is essential for the enhanced coercivity. In our case, due to the weak exchange interaction between NiO and Fe_3O_4, the presence of a NiO under layer marginally increases the coercivity of the Fe_3O_4/NiO bilayer[31] and the AF domain energy in NiO layer results in an asymmetric transversal MR.[32]

Fig. 3 shows the bias dependence of the longitudinal (V_{xx}), transversal voltage (V_{xy}), second order transversal resistance (d^2V_{xy}/dI^2), and transversal MR at 1 T field measured for the Fe_3O_4 single layer Figs. 3(a)–3(d) and for the Fe_3O_4/NiO bilayer Figs. 3(e)–3(h). One can clearly see that the longitudinal voltage presents a linear dependence on the current, while the transversal voltage presents a non-linear dependence. The non-linear dependence of transversal voltage vs bias current may indicate different scattering mechanisms for different bias regions. The resistivity for the Fe_3O_4 single layer is higher than that of Fe_3O_4/NiO bilayer with values of 3.15×10^{-4} and 2.33×10^{-4} Ωm, respectively. The difference in the strain status of Fe_3O_4 layer should not play a role since the thickness of the NiO interlayer (15 nm) is well below the critical thickness for the NiO/MgO system (60 nm),[33] and the NiO layer should remain fully coherent with the MgO(001) substrate. The observed difference can be understood by exchange interaction between Fe_3O_4 and NiO. This interfacial interaction causes pinning of the spin

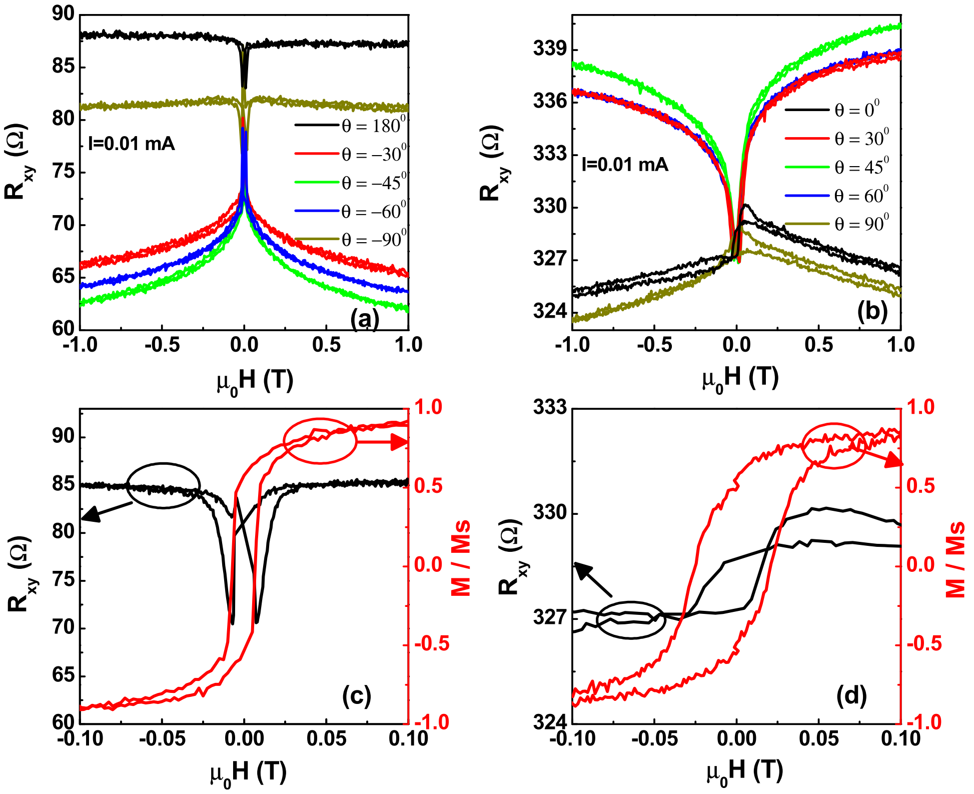

FIG. 2. Transversal MR vs magnetic field at 300 K, measured at different angles for 10 nm Fe_3O_4 (a) and Fe_3O_4/NiO bilayer (b). For comparison with the transversal MR measured at $\theta = 0^0$, magnetization vs field measurements along [100] direction for both systems are also shown in (c) and (d).

chains on both sides of the AF coupled APB. This may reduce the angle between neighboring spins across a single APB, therefore increasing the conductivity as the dependency is to the $\cos^2\theta$ of this angle.[28,34] Interestingly, the transversal MR in both systems strongly depends on current density. One can clearly identify three regions from Figs. 3(d) and 3(h). In region I: the transversal MR ratio is nearly constant. In region II: the transversal MR ratio decreases initially with increasing bias current and exhibits a minimum at a critical bias where the d^2V_{xy}/dI^2 shows a peak. Further increasing the bias increases the transversal MR ratio. In region III: the transversal MR ratio is nearly constant and shows no angular dependence. We would like to point out that the AF exchange interaction between NiO and magnetite can modify the value of critical bias which may benefit the devices application.

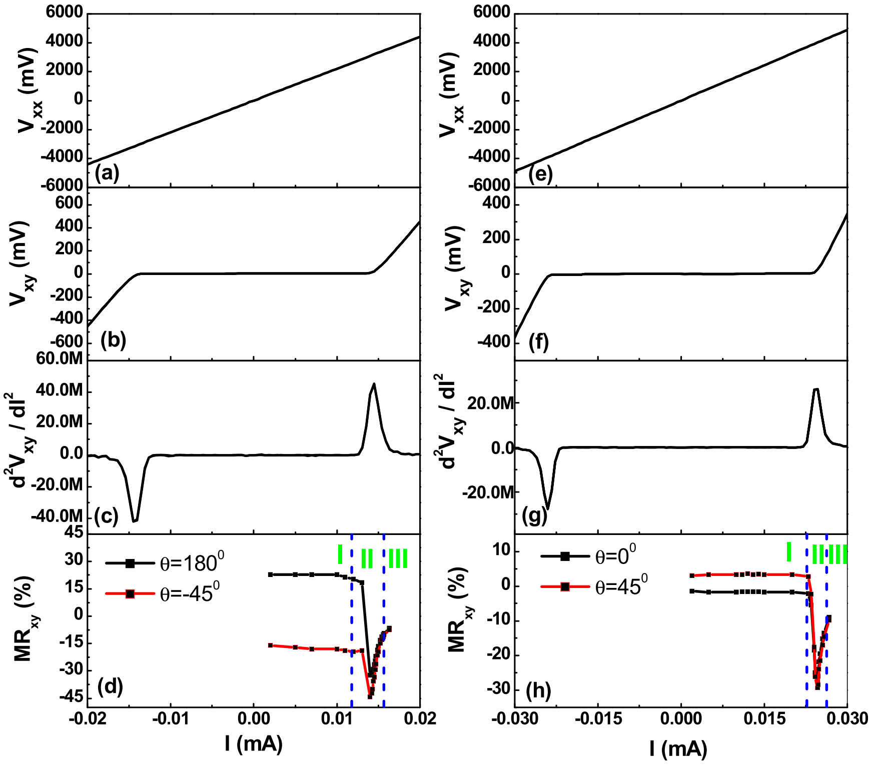

FIG. 3. Bias dependence of the longitudinal (V_{xx}) and transversal voltage (V_{xy}) for both systems under a 1 T magnetic field. (a)-(c) 10 nm Fe_3O_4/MgO, (e)-(g) 10 nm Fe_3O_4/NiO/MgO. The bias dependence of the transversal MR for a magnetic field of 1 Tesla for Fe_3O_4/MgO (d) and 10 nm Fe_3O_4/NiO/MgO (h).

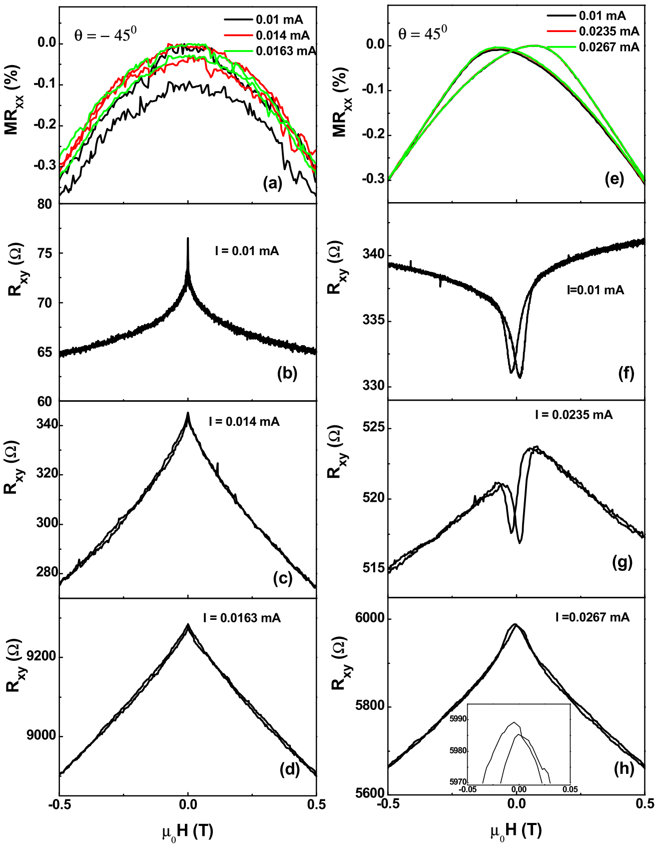

FIG. 4. Magnetic field dependence of the longitudinal and transversal MR measured at three current regions for (a)-(d) Fe_3O_4 film grown on MgO and (d)-(h) Fe_3O_4 film grown on NiO/MgO.

In order to understand the underlying mechanism of transversal MR at different bias current, Fig. 4 shows the magnetic field dependence of the longitudinal and transversal MR measured at different bias regions for Fe_3O_4 (Figs. 4(a)–4(d)) and Fe_3O_4/NiO (Figs. 4(e)–4(h)), respectively. It is interesting to point out that the longitudinal MR is independent of the applied bias current (see Figs. 4(a) and 4(e)). In contrast, one can observe that the transversal MR shows a very clear bias dependence, in which we distinguish 3 regions as described above. It seems that with increasing bias current, there is an increased spin scattering at the APBs which results in a MR with linear dependence vs magnetic field at high magnetic field.[27,28] In region I (Figs. 4(b) and 4(f)), for a small bias current, the spin scattering at the APBs may not play an important role. Thus, the magnetic field dependence of the resistance can be described by the conventional transversal MR for a single magnetic domain.[17] Interestingly, in region II transversal MR with conventional field dependence and the one with linear dependence vs magnetic field coexist which can be clearly seen from Figs. 4(c) and 4(g). At low magnetic field, the transversal MR is dominated by the contributions from the conventional transversal MR. While at high magnetic field, the transversal MR is due to electron spin-dependent transport across antiferromagnetic APBs. Especially, in region III (Figs. 4(d) and 4(h)), the transversal MR shows a linear response with respect to the magnetic field, and the asymmetry with respect to the applied field for NiO underlayer is not as significant as in regions I and II (see inset of Fig. 4(h)). Therefore, the transversal MR in metal oxide with APBs cannot be described by the conventional transversal MR for a single magnetic domain. The effect of electron scattering at the APBs may have to be considered.

The origin of transversal voltage is believed to be closely related to the AMR.[10] Considering the two band model of AMR, the amplitude ($\rho_{||}$-$\rho_{\perp}$) can be shown as[35]

$$\rho_{||} - \rho_{\perp} = \rho_0 \gamma_{eff}(\alpha - 1) \qquad (5)$$

where ρ_0 is the average resistivity, $\alpha = \rho_{\uparrow}/\rho_{\downarrow}$ (where $\rho_{\uparrow}(\rho_{\downarrow})$ is the resistivity experienced by up (down) spin electrons) and γ_{eff} is the parameter which depends upon crystal field splitting, exchange field splitting, spin orbit interaction potential, α and $\beta = \rho_{\uparrow\downarrow}/\rho_{\downarrow}$, where $\rho_{\uparrow\downarrow}$ is the spin flip scattering resistivity. It is known that the largest contribution to resistivity in magnetite films is due to the spin scattering at APBs.[27,28] One can assume the APB scattering is bias

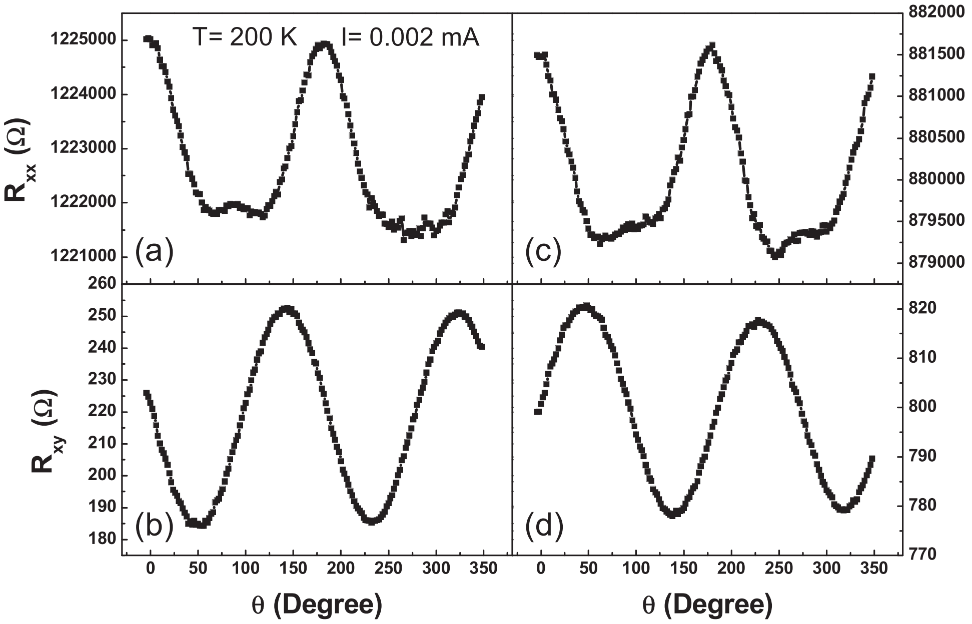

FIG. 5. Longitudinal and transversal MR as a function of θ under 1 T field measured at 200 K for (a) and (b) Fe_3O_4 film grown on MgO and (c) and (d) Fe_3O_4 film grown on NiO/MgO. The bias current is 0.002 mA for both systems.

dependent. Thus, affecting the spin flip scattering coefficient β will in turn affect the MR amplitude ($\rho_{||}$-$\rho_{\perp}$) according to Eq. (5).[8,34] If this is the case, Eqs. (1) and (2) suggest that the influence of spin scattering events on AMR and transversal MR should be equal, but our results show that the bias dependence is only affecting the transversal MR, suggesting that a more rigorous theory is needed to explain the transversal MR. It is known that additional set of peaks appear in AMR in magnetite when the temperature is below 250 K.[6] This was explained by the onset of the charge ordering once the temperature approaches Verwey temperature. Fig. 5 shows the longitudinal and transversal MR as a function of θ under 1 T field measured at 200 K. One can see that an additional peak is present for AMR measurements but not for transversal MR at low bias which may indicate that the charge ordering may not affect the transversal MR in magnetite. As low bias transversal MR in magnetite can be described by conventional transversal MR, our results may also suggest that the current explanation of the origin of transversal MR on the basis of anisotropy MR alone may not be sufficient for a system experiencing charge ordering.

In summary, the magnetization reversal process in Fe_3O_4 layer and Fe_3O_4/NiO bilayer was examined by magnetotransport measurements. The transversal MR shows dependence with the applied bias current where three different regions can be identified. This suggests that care must be taken when evaluating the conventional transversal MR on magnetite. Our analysis suggests that the transversal MR in magnetite cannot be explained by the conventional transversal MR in the single domain case and the existing theory based on AMR alone is insufficient to explain the transversal MR in magnetite. Moreover, the observed bias dependence can be utilized for the improved performance of transversal MR devices.

This work was supported by the Science Foundation of Ireland (SFI) under Contract No. 06/IN.1/I91, 08/ERA/I1760. H.C.W. is grateful to the SFI Short Term Travel fellowship support during his stay at PKU and thanks Professor O. N. Mryasov at MINT for useful discussion.

[1] I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).

[2] B. Dieny, V. S. Speriosu, S. S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B **43**, 1297 (1991).

[3] F. Montaigne, A. Schuhl, F. N. Van Dau, and A. Encinas, Sens. Actuators, A **81**, 324 (2000).

[4] D. L. Graham, H. A. Ferreira, and P. P. Freitas, Trends Biotechnol. **22**, 455 (2004).

[5] A. Schuhl, F. N. Van Dau, and J. R. Childress, Appl. Phys. Lett. **66**, 2751 (1995).

[6] R. Ramos, S. K. Arora, and I. V. Shvets, Phys. Rev. B **78**, 214402 (2008).

[7] K. M. Seemann, F. Freimuth, H. Zhang, S. Blügel, Y. Mokrousov, D. E. Bürgler, and C. M. Schneider, Phys. Rev. Lett. **107**, 086603 (2011).

[8] N. Naftalis, A. Kaplan, M. Schultz, C. A. F. Vaz, J. A. Moyer, C. H. Ahn, and L. Klein, Phys. Rev. B **84**, 094441 (2011).

[9] Y. Bason, L. Klein, J.-B. Yau, X. Hong, and C. H. Ahn, Appl. Phys. Lett. **84**, 2593 (2004).

[10] T. McGuire and R. Potter, IEEE Trans. Magn. **11**, 1018 (1975).

[11] W. Döring, Ann. Phys. **424**, 259 (1938).

[12] Y. Bason, J. Hoffman, C. H. Ahn, and L. Klein, Phys. Rev. B **79**, 092406 (2009).

[13] N. Naftalis, Y. Bason, J. Hoffman, X. Hong, C. H. Ahn, and L. Klein, J. Appl. Phys. **106**, 023916 (2009).

[14] G. Li, T. Yang, Q. Hu, H. Jiang, and W. Lai, Phys. Rev. B **65**, 134421 (2002).

[15] L. Ejsing, M. Hansen, A. Menon, H. Ferreira, D. Graham, and P. Freitas, Appl. Phys. Lett. **84**, 4729 (2004).

[16] Y. Bason, L. Klein, J. Yau, X. Hong, J. Hoffman, and C. H. Ahn, J. Appl. Phys. **99**, 08R701 (2006).

[17] X. Jin, R. Ramos, Y. Zhou, C. McEvoy, and I. V. Shvets, J. Appl. Phys. **99**, 08C509 (2006).

[18] A. Fernández-Pacheco, J. M. De Teresa, J. Orna, L. Morellon, P. A. Algarabel, J. A. Pardo, M. R. Ibarra, C. Magen, and E. Snoeck, Phys. Rev. B **78**, 212402 (2008).

[19] S. U. Jen, P. J. Wang, Y. C. Tseng, and H. P. Chiang, J. Appl. Phys. **105**, 07E903 (2009).

[20] K. Ghosh, S. Ogale, S. Pai, M. Robson, E. Li, I. Jin, Z. Dong, R. Greene, R. Ramesh, T. Venkatesan, and M. Johnson, Appl. Phys. Lett. **73**, 689 (1998).

[21] P. Seneor, A. Fert, J. Maurice, F. Montaigne, F. Petroff, and A. Vaurès, Appl. Phys. Lett. **74**, 4017 (1999).

[22] X. Li, A. Gupta, G. Xiao, W. Qian, and V. Dravid, Appl. Phys. Lett. **73**, 3282 (1998).

[23] H. C. Wu, S. K. Arora, O. N. Mryasov, and I. V. Shvets, Appl. Phys. Lett. **92**, 182502 (2008).

[24] W. Eerenstein, T. T. M. Palstra, T. Hibma, and S. Celotto, Phys. Rev. B **66**, 201101(R) (2002).

[25] L. McGuigan, R. C. Barklie R. G. S. Sofin, S. K. Arora, and I. V. Shvets, Phys. Rev. B **77**, 174424 (2008).

[26] S. K. Arora, H. C. Wu, R. J. Choudhary, I. V. Shvets, O. N. Mryasov, H. Z. Yao, and W. Y. Ching, Phys. Rev. B **77**, 134443 (2008).

[27] W. Eerenstein, T. T. M. Palstra, S. S. Saxena, and T. Hibma, Phys. Rev. Lett. **88**, 247204 (2002).

[28] H. C. Wu, M. Abid, B. S. Chun, R. Ramos, O. N. Mryasov, and I. V. Shvets, Nano Lett. **10**, 1132 (2010).

[29] H. Yang, S. H. Yang, D. C. Qi, A. Rusydi, H. Kawai, M. Saeys, T. Leo, D. J. Smith, and S. S. P. Parkin, Phys. Rev. Lett. **106**, 167201 (2011).

[30] C. Gatel, E. Snoeck, V. Serin, and A. R. Fert, Eur. Phys. J. B **45**, 157 (2005).

[31] Z. Li and S. Zhang, Phys. Rev. B **61**, R14897 (2000).

[32] Z. Q. Lu, G. Pan, and W. Y. Lai, J. Appl. Phys. **90**, 1414 (2001).

[33] M. A. James and T. Hibma, Surf. Sci. **433**, 718 (1999).

[34] R. G. S. Sofin, H. C. Wu, and I. V. Shvets, Phys. Rev. B **84**, 212403 (2011).

[35] M. Ziese, Phys. Rev. B **62**, 1044 (2000).