https://doi.org/10.30799/jnst.166.18040518 J. Nanosci. Tech. - Volume 4 Issue 5 (2018) 533–535

Share Your Innovations through JACS Directory

Journal of Nanoscience and Technology

Visit Journal at http://www.jacsdirectory.com/jnst

ISSN: 2455-0191

Influence of Miscut Angle on the Magnetoresistance Behavior of Epitaxial Fe_3O_4 Thin Films Grown on Vicinal Substrates

Sunil Kumar Arora[1],*, Igor V. Shvets[2]

[1]*Centre for Nano Science & Nano Technology, South Campus, Panjab University, Sector 25, Chandigarh – 160 014, Punjab, India.*
[2]*School of Physics, Trinity College Dublin, Dublin – 2, Ireland.*

ARTICLE DETAILS

Article history:
Received 02 October 2018
Accepted 23 October 2018
Available online 11 November 2018

Keywords:
Vicinal Surface
Magneto Resistance
Spin-Electronics

ABSTRACT

This present work reported here the influence of miscut angle of vicinal MgO (100) substrates (with miscut angle between 1-5° along <011> direction) on the magnetoresistance (MR) behavior of epitaxial magnetite (Fe_3O_4) films. By varying the miscut angle, we are able to control the step-terrace periodicity of the substrate surface, which allows us to tailor the density of the step-edges induced antiphase boundaries, APBs (nanoscale liner defects formed during growth due to coalescence of neighbouring domains), in epitaxial Fe_3O_4 films. We found that the contribution from the step-edges induced APBs to the magnetoresistance (MR_{SE}) in Fe_3O_4 films increases with increase in miscut (up to 2°) and decreases for larger miscut angles. From temperature dependent studies, we observed that the magnitude of MR_{SE} increased with decrease in temperature and peaks at the Verwey transition. The decrease in MR_{SE} for larger miscut samples is attributed to the influence of APB strain-fields on the degree of spin-polarization of Fe_3O_4 regions on terraces.

1. Introduction

Recently, oxide heterostructures have attracted a lot of attention as they display a vast range of physical phenomena like conductivity, magnetism, or even superconductivity [1-7]. In most cases, these effects are caused by electron correlations and are also interesting for studying fundamental physics. Many of the observed phenomena are affected by the presence of defects (grown in and interfacial defects) [8-10]. This is due to the fact that any disturbance to the crystal structure is liable to significantly alter properties such as conductivity and magnetic ordering. Altered local crystal structure and symmetry affects the electron hopping mechanisms and super exchange interactions. In previous studies efforts have been made to manipulate the defect density at the interface and in the overlayer by tailoring the surface topography of substrate. In fact by appropriate control of the surface topography has been demonstrated to greatly influence the structure and properties of the interfaces and thin films [1,9,11,12].

Many oxide based heterostructures have been studied which includes half metallic oxides (Fe_3O_4, CrO_2, manganites etc.) and other oxides (NiO, MgO, $SrTiO_3$, $LaAlO_3$ etc.) [1-12]. Fe_3O_4 is highly ranked as a possible candidate to be used as spin electrode in the field of spintronic owing to its high Curie temperature and half metallic nature [7,11,15]. Fe_3O_4 has been grown successfully on variety of substrates using pulsed laser deposition and molecular beam epitaxy techniques. Commonly used subsrtae for epitaxial growth of Fe_3O_4 is MgO (100) [7, 11-14]. The Fe_3O_4-MgO (100) heteroepitaxial system suffer from the formation of structurally shifted domains called antiphase boundaries (APBs). The APBs are formed as a natural growth defect due to differences in the translational and rotational symmetry between Fe_3O_4 (*Fd3m*) and MgO (*Fm3m*). Typically in a Fe_3O_4-MgO heteroepitaxial system, the APBs will form at random locations and lead to unsaturated or reduced magnetization (due to antiferromagnetic exchange at the boundary) [16] and enhanced magnetoresistance (spin polarized tunneling across the boundary) [17]. In our previous study we have shown that by exploiting the surface topography of vicinal MgO (100) substrates (which are miscut from the low index plane along <011> direction) one can intentionally induce the APBs in Fe_3O_4 and arrange them in a parallel array to obtain enhanced magnetoresistance [11]. The layered inverse spinel crystal structure of Fe_3O_4 and step-terrace periodic structures of the MgO (100) substrate (with miscut either along <001> or <011> direction) facilitate the formation of structurally shifted domains (APBs) in the growth direction. In another report Sofin et al. [18] reported a large anisotropy in the MR in Fe_3O_4 films on MgO (011). They claimed that the difference in number of APB defects produced along <011> and <001> directions was responsible for the observed MR anisotropy. Here, we present a systematic study to understand the influence APB density on the magnetoresistance behavior of Fe_3O_4-MgO heteroepitaxial system. The density of APBs in Fe_3O_4-MgO heteroepitaxial system is controlled by manipulating surface step-terrace periodicity through the miscut angle of MgO(100) substrates which have miscut along the <011> direction. The observed dependency of MR on miscut angle and temperature are described on the basis of spin dependent scattering across the atomically sharp interfaces of APBs and influence of strain on the spin polarization of the film region in its vicinity.

2. Experimental Methods

Epitaxial Fe_3O_4 films used in the present study are of 45 nm thickness and grown on MgO (100) substrates using an oxygen plasma assisted molecular beam epitaxy (MBE) system having a base pressure of 5×10^{-10} Torr (DCA MBE600). The MgO substrates had a varying degree of miscut (1 to 5°) along the <011> direction. Prior to film growth the substrate surface was pretreated with an optimized annealing sequence to obtain the desired step-terrace periodic structure, details of which are described in a previous report [12]. The growth was carried out at a substrate temperature of 250 °C. We grew four samples of epitaxial Fe_3O_4 using the optimized growth conditions on vicinal MgO (100) substrate having miscut of 1, 2, 3.4 and 5 degrees along the <011> direction. In situ reflection high-energy electron diffraction (RHEED) was used to determine the growth rate and growth mode (STAIB). It was also used to determine the extent of miscut as well as step terrace periodicity. Details of the film growth procedure are given elsewhere [12]. The X-ray rocking curves for the symmetric (002) and asymmetric (311) Bragg reflections was performed using a high resolution X-ray diffractometer (Bede-D1) operated in the triple-axis configuration. The field and temperature dependence of electrical resistivity and magnetoresistance (MR) were performed in the standard dc-four probe technique using a physical property measurement system(PPMS) equipped with a 14 T

---

*Corresponding Author:aroras@pu.ac.in(Sunil Kumar Arora)

https://doi.org/10.30799/jnst.166.18040518

Cite this Article as: Sunil Kumar Arora, Igor V. Shvets, Influence of miscut angle on the magnetoresistance behavior of epitaxial Fe_3O_4 thin films grown on vicinal substrates, J. Nanosci. Tech. 4(5) (2018) 533–535.

superconducting magnet (PPMS 6000, Quantum Design). The sample rotator probe on the PPMS system facilitates orientations dependence studies. The MR is defined as $MR(\%) = \frac{R(H)-R(0)}{R(0)} \times 100$, where $R(H)$ and $R(0)$ are the resistances of the sample with and without field (H), respectively. The MR results reported here are obtained by keeping the direction of magnetic field in-plane and parallel to the current direction. To determine the MR anisotropy, the measurements have been carried out by passing current in two directions, which are orthogonal to each other, i.e., along- and perpendicular- to the direction of step-edges (SE) denoted as SE and PSE respectively.

3. Results and Discussion

Prior to discussing the results of magneto-transport studies and their correlation with the APBs density, we would like to mention that in our previous studies we have reported in detail the procedure to obtain regular step-terrace periodicity on vicinal MgO (100) substrates [12]. The procedure involves annealing the substrate under UHV conditions followed by high temperature anneal in the presence of oxygen plasma. The step terrace periodicity of the substrate and film was confirmed from in-situ RHEED. Through detailed high resolution cross-sectional TEM investigations, we observed that the films possess the epitaxial relationship with the substrate and step-terrace periodicity propagated through the entire thickness [11, 14]. Magnetization studies of Fe_3O_4 films on vicinal MgO revealed an in-plane uniaxial magnetic anisotropy induced by step-edges with an easy axis along the PSE direction for temperatures above the Verwey transition [19]. Whereas, for temperatures below the Verwey transition there was a transition of easy axis from PSE to SE, which was also found to be thickness dependent. We further discussed the possibility of formation of APBs related to the shift vectors in Fe_3O_4 layers on stepped surface and related the observed magnetic anisotropy to the altered magnetic interactions at the APBs [19].

The ω-2θ rocking curves measured at room temperature for (200) and (400) Bragg reflections of the MgO and Fe_3O_4 respectively for Fe_3O_4 thin films grown on 1 and 2 degree miscut substrates are shown in Fig. 1. The horizontal axis in the figure is shown with reference to the Bragg angle for symmetric (200) reflection of MgO substrate. The out-of-plane lattice constant determined for both the films is found to be 0.8372 nm. The full width at half maximum (FWHM) for the thin film peak is found to be 0.064 and 0.079 degrees for 1 and 2 degree miscut samples. For higher miscut samples out-of-plane lattice constant was found to be same (0.8372 nm) and the FWHM was comparable to that of 2 degree miscut sample. Larger FWHM of the thin film peak for samples with 2 degree and higher miscut samples represents additional scattering contribution arising from the presence of step edges. The asymmetric (622)/(311) Bragg reflections (not shown) measured for all the samples were used to determine the in-plane lattice constant of Fe_3O_4 thin films and found to be 0.84236 (±0.00004) nm, which is twice that of the MgO substrate (0.4213 nm). This confirms the Fe_3O_4 films grow epitaxially and maintain one-to-one registry with the MgO substrate.

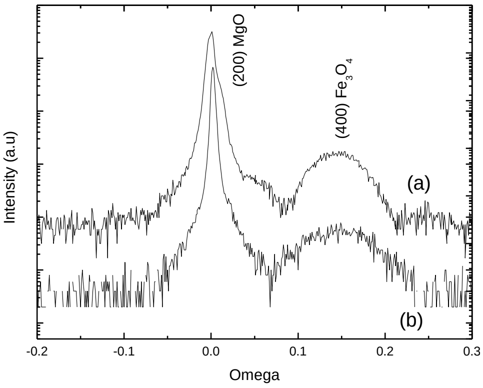

Fig. 1 The ω–2θ_ rocking curves of Fe_3O_4 films on 1 and 2 degree miscut MgO (100) substrates (curves a and b respectively) measured at room temperature and relative to the (200) Bragg reflection of MgO

Prior to discussing the MR results, we would like to mention that from the analysis of the temperature dependence of resistivity for the sample grown on 1, 2, 3.4 and 5 degrees miscut vicinal MgO substrates, we found that the Verwey transition temperature (T_v) is not affected by the direction of current flow with respect to miscut. This is in line with the previously reported results [11, 19]. Value of the T_v for current parallel to SE are found to be 110.5, 110, 109, 107 K respectively. However, the resistivity was found to be higher when measured across SE by 6, 8, 12, 15% as compared to resistivity value measured along the SE for 1, 2 3.4 and 5 degree miscut respectively. The activation energies for the region above and below the T_v were found to be in the range of 30-35 meV and 55-65 meV. Fig. 2 shows a representative MR curve as a function of magnetic field (H) measured for both SE and PSE directions at 300, 130, 110 and 100 K for the Fe_3O_4 sample grown on MgO (100) substrate with 1° miscut along <011>. Magnitude of MR for both directions is found to increase with the decrease in temperature and peaks at the Verwey transition temperature (110 K). One also notices that the MR has a sharper field dependence at lower magnetic fields (smaller than 0.5 T) than at higher magnetic fields. This feature is more prominent at lower temperatures for PSE direction. Temperature dependence of MR at 2 Tesla field for both the directions for the same sample is shown in Fig. 3. The maximum value of MR (2T) is found to be 10.4% at the T_v (110 K) for current flow in the SE direction and is 4.6% greater than the corresponding MR value for SE direction.

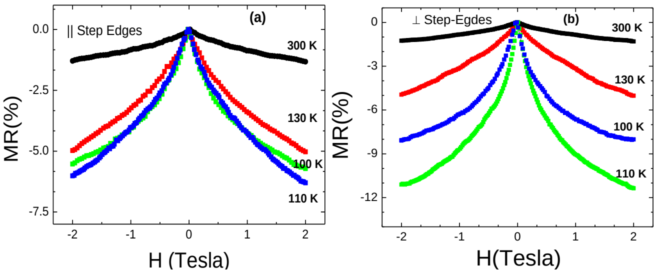

Fig. 2 Magnetoresistance (MR) as a function of applied in-plane magnetic field (H) for a 45 nm thick epitaxial Fe_3O_4 films on 1° miscut MgO (100) substrate measured at different temperatures with current direction along (a) SE and (b) PSE directions. The direction of current and magnetic field are parallel to each other in both cases

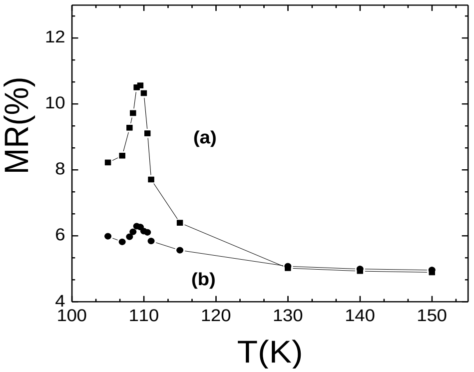

Fig. 3 Temperature dependence of MR at 2 Tesla field measured along (a) PSE and (b) SE directions for a 45 nm thick epitaxial Fe_3O_4 films on MgO (100) substrate having 1° miscut along the <011>

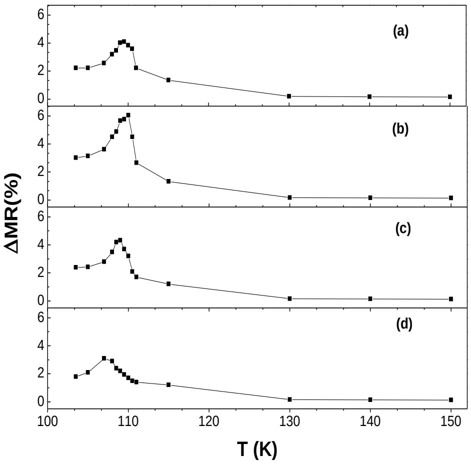

Fig. 4 Temperature dependence of MR_{SE} (step-edge induced MR) measured for all the four samples having a miscut magnitude of (a) 1°, (b) 2°, (c) 3.4° and (d) 5°

The difference in MR between the two directions (MR_{SE}) represents the MR contribution arising from the step-edges induced APBs. The MR_{SE} as a function of T is shown in Fig. 4 for all the four samples which are having different degree of miscut. One notices that the MR_{SE} peaks at T_v and its magnitude is maximum for the 2 degree miscut (6.5% at 110 K). For samples with higher miscut of 3.4 and 5 degrees, magnitude of MR and MR_{SE} are decreased.

https://doi.org/10.30799/jnst.166.18040518

Cite this Article as: Sunil Kumar Arora, Igor V. Shvets, Influence of miscut angle on the magnetoresistance behavior of epitaxial Fe_3O_4 thin films grown on vicinal substrates, J. Nanosci. Tech. 4(5) (2018) 533–535.

The observed changes in MR as a function of miscut magnitude and temperature can be qualitatively understood from the fact that in our case, due to the presence of step-terrace periodic pattern with atomic height steps (0.21 nm) on the MgO (100) surface, out-of-plane shifted APBs are formed leading to additional spin scattering form the local structural and spin disorder [19]. Presence of APBs modifies the transfer integral t_{ij} that governs the electronic transport between Fe^{2+} and Fe^{3+} octahedral cations [20, 21]. These periodic perturbations in the electronic structure at the step-edges produce greater contribution of spin dependent scattering for current flow along PSE than SE (parallel to terraces). Furthermore, the strain field of the APBs alters (reduces) the degree of spin polarization of the film region in its vicinity causing MR to decrease for both the directions. The peak in MR was observed in the vicinity of the T_V results from abrupt changes in the thermodynamic quantities accompanied with the first order Verwey transition [20, 21].

To explain the MR_{SE} dependence on miscut angle, one can consider that with the increase in miscut, number of APBs formed by step-edges will increase, leading to an enhanced MR_{SE}. This explains the observed T dependence of MR_{SE} for miscut angles of upto 2 degrees. The observed decrease in MR_{SE} for larger miscut angles (>2 degrees), can be related to the reduction in the terrace width with increase in miscut and subsequent effect of strain fields associated with the APB on the Fe_3O_4 regions in its vicinity. The estimated value of terrace width for miscut angles of 1, 2, 3.4 and 5 degree miscut are 8.4, 4.2, 2.52 and 1.68 nm respectively. As the terrace width is only 2 and 3 unit cells of Fe_3O_4 for 3.4 and 5 degree miscut samples respectively, the strain fields and modified cationic configuration at the SE induced APBs will affect (reduce) the degree of spin-polarization as well as magnetic exchange of Fe_3O_4 region in its vicinity [7, 20, 21]. This in turn will produce greater proportion of regions of reduced spin polarization leading to a reduced MR for both the directions and lowering of MR_{SE} magnitude throughout the temperature range.

4. Conclusion

The results conclude that the step-edge induced APBs in Fe_3O_4 film on vicinal MgO (100) substrates influences the spin-transport behaviour significantly. The contribution to MR arising due to the step-edges induced scattering is found increased with the increase in miscut angle (step-density) and is optimum for the 2 degree miscut sample. The MR_{SE} is also found to be temperature dependent with its magnitude being maximum at the T_v. The observed decrease in magnitude of MR and MR_{SE} for larger miscut (>2 degrees) samples is related to the strain fields experienced by the Fe_3O_4 regions on the terraces originating from the step-edge induced APBs, which cause the spin-polarization of Fe_3O_4 regions to reduce leading to a lower magnitude of MR and decrease of MR_{SE}.

Acknowledgements

The financial support from the Science Foundation Ireland is gratefully acknowledged (under contract no. 06/IN.1/191). The authors also would like to thank the DST for financial support under the PURSE grant.

References

[1] J. Chakhalian, J.W. Freeland, A.J. Millis, C. Panagopoulos, J.M. Rondinelli, Colloquium: Emergent properties in plane view: Strong correlations at oxide interfaces, Rev. Mod. Phys. 86 (2014) 1189-1202.

[2] H. Chen, A. Millis, Charge transfer driven emergent phenomena in oxide heterostructures, J. Phys. Cond. Matter 29 (2017) 243001-1-7.

[3] H.Y. Hwang, Y. Iwasa, M. Kawasaki, B. Keimer, N. Nagaosa, Y. Tokura, Emergent phenomena at oxide interfaces, Nature Mater. 11(2) (2012) 103-113.

[4] G.N. Daptary, S. Kumar, A. Bid, P. Kumar, A. Dogra, R.C. Budhani, D. Kumar, N. Mohanta, A. Taraphder, Observation of transient superconductivity at the $LaAlO_3/SrTiO_3$ interface, Phys. Rev. B 95 (2017) 174502-1-10.

[5] T.L. Kim, H.W. Jang, Tailoring two-dimensional electron gas conductivity at oxide heterointerfaces, Curr. Appl. Phys. 17 (2017) 626-639.

[6] F. Bi, M. Huang, H. Lee, C.B. Eom, P. Irvin, J. Levy, $LaAlO_3$ thickness window for electronically controlled magnetism at $LaAlO_3/SrTiO_3$ heterointerfaces, Appl. Phys. Lett. 107 (2015) 082402-1-4.

[7] S.K. Arora, H.C. Wu, R.J. Choudhary, I.V. Shvets, O.N. Mryasov, H.Z. Yao, W.Y. Ching, Giant magnetic moment in epitaxial Fe_3O_4 thin films on MgO(100), Phys. Rev. B 77 (2008) 134443-1-5.

[8] N. Nakagawa, H.Y. Hwang, D.A. Muller, Why some interfaces cannot be sharp, Nature Mater. 5 (2006) 204-209.

[9] J.A. Sulpizio, S. Ilani, P. Irvin, J. Levy, Nanoscale phenomena in oxide heterostructures, Annual Rev. Mater. Res. 44 (2014) 117-149.

[10] Z. Wang, Z. Zhong, S.M. Walker, Z. Ristic, J.Z. Ma, F.Y. Bruno, et al., Atomically precise lateral modulation of a two-dimensional electron liquid in anatase TiO_2 thin films, Nano Lett. 17 (2017) 2561-2567.

[11] S.K. Arora, R.G.S. Sofin, I.V. Shvets, Magnetoresistance enhancement in epitaxial magnetite films grown on vicinal substrates, Phys. Rev. B 72 (2005) 134404-1-10.

[12] R.G.S. Sofin, S.K. Arora, Shvets, Influence of substrate pre-deposition annealing on step edges-induced magnetoresistance in epitaxial magnetite films grown on vicinal MgO (100) substrates, Proceedings of the Joint European Magnetic Symposia, San Sebastian, Spain, J. Magn. Magn. Mater. 316 (2007) E969- E 972.

[13] Ramos, S.K. Arora, I.V. Shvets, Influence of miscut on the anisotropic magnetoresistance of magnetite thin films, J. Appl. Phys. 105 (2009) 07B108-1-3.

[14] H.C. Wu, X. Liu, C. Coileain, H. Xu, M. Abid, et al., Competition between anti-phase boundaries and charge-orbital ordering in epitaxial stepped Fe_3O_4 (100) thin films, Spin 7 (2017) 1750001-1-8.

[15] Zutic, J. Fabian, S. Das Sarma, Spintronics: Fundamentals and applications, Rev. Mod. Phys. 76 (2004) 323-410.

[16] D.T. Margulies, F.T. Parker, M.L. Rudee, F.E. Spada, J.N. Chapman, P.R. Aitchison, A.E. Berkowitz, Origin of the anomalous magnetic behavior in single crystal Fe_3O_4 films, Phys. Rev. Lett. 79 (1997) 5162-5165.

[17] W. Eerenstein, T. Hibma, S. Celotto, Mechanism for superparamagnetic behavior in epitaxial Fe_3O_4 films, Phys. Rev. B 70 (2004) 184404-1-6.

[18] R.G.S. Sofin, S.K. Arora, I.V. Shvets, Positive antiphase boundary domain wall magnetoresistance in Fe_3O_4 (110) heteroepitaxial films, Phys. Rev. B 83 (2011) 134436-1-9.

[19] H.C. Wu, A. Syrlybekov, O. Mauit, A. Mouti, C. Coileáin, et al., Magnetic and transport properties of epitaxial stepped Fe_3O_4 (100) thin films, Appl. Phys. Lett. 105 (2014) 132408-1-3.

[20] H.T. Jeng, G.Y. Guo, First-principles investigations of the electronic structure and magnetocrystalline anisotropy in strained magnetite Fe_3O_4, Phys. Rev. B 65 (2002) 094429-1-9.

[21] Z. Zhang, S. Satpathy, Electron states, magnetism, and the Verwey transition in magnetite, Phys. Rev. B 44 (1991) 13319-13331.

https://doi.org/10.30799/jnst.166.18040518

Cite this Article as: Sunil Kumar Arora, Igor V. Shvets, Influence of miscut angle on the magnetoresistance behavior of epitaxial Fe_3O_4 thin films grown on vicinal substrates, J. Nanosci. Tech. 4(5) (2018) 533–535.