Asymmetric Magnetization Reversal Probed by Recoil Loop Measurements in an Exchange Biased $La_{0.67}Sr_{0.33}MnO_3/La_{0.33}Ca_{0.67}MnO_3$ Bilayer Film

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We exploit the recoil loop measurements to study the asymmetric magnetization reversal in an exchange-biased $La_{0.37}Sr_{0.33}MnO_3/La_{0.33}Ca_{0.67}MnO_3$ bilayer film. It is found that the recoil curve encloses a marked area only in the second quadrant of the hysteresis loop, and the recoil susceptibility in the descending branch of the major loop is evidently higher than that in the ascending branch. The study indicates that the exchange anisotropy of a unidirectional nature and an orientation deviated from the easy axis of the ferromagnetic layer plays a crucial role in creating the reversal asymmetry.

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Interactions between different magnetic phases in a hybrid structure give rise to intriguing magnetic behaviour. In an antiferromagnetic (AF)–ferromagnetic (FM) bilayer film, the exchange coupling across the interface can displace the hysteresis loop along the magnetic field axis. Although this so-called exchange bias phenomenon was discovered half a century ago,[1] it has attracted increasing interest because of its elusive microscopic origin and its technological applications to magnetic read heads and magnetic nonvolatile memories.[2,3] In the past decades, many interesting properties were found in exchange biased systems. Among the enriched connotation of exchange bias, a fundamental issue, which has received considerable attention, is the asymmetry exhibiting in the magnetization reversal process.[4−19]

It has been observed that in the ascending branch of a hysteresis loop, the reversal proceeds by magnetization rotation, in the descending branch it can take place by domain wall nucleation and motion.[5,9,13] Dissimilar scenarios were also reported.[4,12,15,17,19] Experimentally, the reversal asymmetry has been explored by conventional magnetometry, polarized neutron reflectometry,[5,9,16] anisotropic magnetoresistance,[6,10] and magnetic domain imaging techniques.[4,13,14,18] For the convenient magnetometry methodology, the reversal asymmetry was usually revealed by the visible asymmetric shape of the hysteresis loop,[9,11,15] or further by different magnetic viscosities close to the left and right coercive fields, respectively.[7,8]

In this Letter, we propose a new approach, i.e. using recoil loops to study the asymmetry of magnetization reversal. Recoil loop refers to a type of minor loop which results from the removal and reapplication of a demagnetizing field applied to a magnetically saturated material.[20] It is often used to characterize the technical performance such as reversible and irreversible contributions in the demagnetization for a permanent magnet at a given applied field. In recent years, recoil loops are mostly studied in exchange spring magnets that consist of suitably dispersed soft and hard magnetic phases for achieving high values of maximum energy product $(BH)_{\max}$.[21−26] It has been shown that the features of a recoil loop such as its openness or its eye effect, may have a close correlation with the inter-granular or inter-layer exchange coupling between the soft and hard magnetic phases. In the present work, the studied material is an exchange biased $La_{0.67}Sr_{0.33}MnO_3$ (LSMO, FM)/$La_{0.33}Ca_{0.67}MnO_3$ (LCMO, AF) bilayer film. By measurements of major and minor loops, we aim to find whether the recoil loops can be exploited to manifest the asymmetric magnetization reversal, and consequently, can provide insights to the interfacial AF–FM exchange coupling.

Bilayer films LSMO (t = 15 nm)/LCMO (t = 30 nm) was grown onto single crystal $SrTiO_3$(STO) (100) substrate by pulsed laser deposition using a condition reported elsewhere.[27] The $\theta - 2\theta$ x-ray scans reveal a crystallographic orientation of $(00l)$ for both AF and FM layers. It is worth noting that the first epitaxial growth of the FM-LSMO layer then covered by an AF-LCMO layer is based on the lattice mismatch between the film materials and STO. The lattice constant of cubic STO is 3.905 Å. In the simple pseudocubic description the lattice constants of LSMO and LCMO are 3.88 Å and 3.80 Å, respectively. Therefore, the inverse architecture induces a tensile strain between the layers, and this ensures a usual magne-

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tization configuration with the magnetization vector $\boldsymbol{M}$ lying in the film plane.

The magnetization measurements were carried out using a superconductor quantum interference device magnetometer (Quantum Design, MPMS-7). The small remanent magnetic field ($\sim$10 Oe) of the superconductor magnet was carefully recorded and corrected by measuring the hysteresis loop of a palladium specimen. In order to establish the exchange bias, the bilayer film was cooled from 300 K (above the Néel temperature $\sim$170 K for LCMO) to low temperatures in an applied magnetic field ($H = 1500$ Oe) along the [110] direction.

Representative hysteresis loops measured at 5 and 75 K are shown in Fig. 1. Because the cubic crystallographic [110] is a biaxial magnetic easy-axis of the FM LSMO film grown on STO (100),[28] the measured hysteresis loops exhibit an easy-axis behaviour with a full magnetic remanence. At 5 K, the loop displays a pronounced shift, which in terms of exchange biased field is $H_{eb} = -(H_{c1} + H_{c2})/2 = 63$ Oe ($H_{c1,2}$ are the coercivity, at which $M = 0$ in the descending and ascending branches, respectively). Meanwhile, the coercive field $H_c = (H_{c2} - H_{c1})/2 = 263$ Oe, which is considerably enhanced compared to that (< 50 Oe) of the single layer LSMO film grown onto STO (100).[28] Though sharp irreversible transitions occur in the vicinity of H_{c1} and H_{c2}, the $M-H$ curve appears more rounded at the shoulder in the descending branch than that in the ascending branch. Applying a large cooling field of 50 kOe does not change this asymmetry. These facts indicate that the reversal asymmetry is not associated with an unsaturated state of the FM layer, and for the present bilayer film the magnetization reversal may be dominated by the domain wall propagation in the ascending branch, whereas it is incorporated with an incoherent process in the descending branch. With increasing temperature to 75 K, the loop shift is rather small ($\sim$7 Oe) while no apparent asymmetry in the hysteresis loop can be distinguished (see Fig. 1(b)). This suggests that the observed asymmetry should be caused by the developed exchange anisotropy at the AF–FM interface.

Considering the major loop asymmetry apparently exhibits at 5 K, we measured a series of recoil loops at this temperature. The field cooling procedure before the recoil loop measurements was exactly the same as used for the major loop measurements. Because the magnetic field range can conspicuously affect the features of the minor loops, all recoil loops were measured from the major loop to the centre field at $H = H_{eb}$ and then back to the major loop. The results are illustrated in Fig. 2. It can be seen that at the applied demagnetizing fields all recoil curves deviate from the major hysteresis loop. At the reverse field $H = -290$ Oe in the descending branch, the recoil curves bears little hysteresis. When the reverse fields are approaching H_{c1}, the recoil loops are clearly opened up, exhibiting an eye-like shape. This indicates that a portion of the bilayer film is being driven around a minor hysteresis loop during the recoil process. As the reverse field continues to increase, the recoil loops display no apparent splitting. In contrast, for the ascending branch, the recoil loops all enclose almost zero hysteresis at the reverse fields no matter close to or away from H_{c2}.

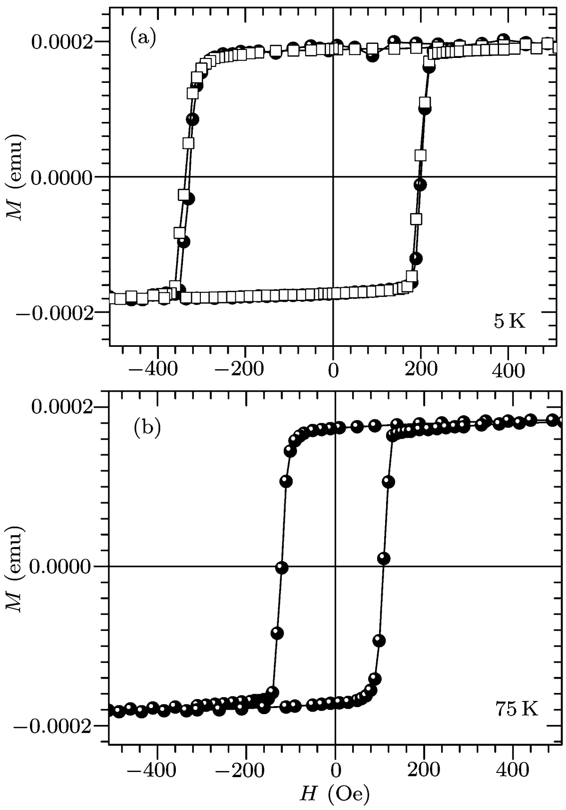

Fig. 1. Hysteresis loops measured at 5 K (a) and 75 K (b). The solid and open symbols represent the data obtained from field cooling in 1.5 kOe and 50 kOe, respectively.

We have compared the recoil susceptibilities (χ) in the ascending and descending branches by measuring the slope of the line connecting the two ends of each recoil loop. As shown in Fig. 3, a hump close to the corresponding coercive fields occurs in the descending and ascending branches. It is obvious that the peak value in the former branch is much higher than that in the latter. Hence, the above results clearly demonstrate that the asymmetric magnetization reversal can be characterized by the recoil loop measurements.

For shedding lights on the exchange spring process in nano-scaled permanent magnetic materials, recoil loops have also been studied by computational means.

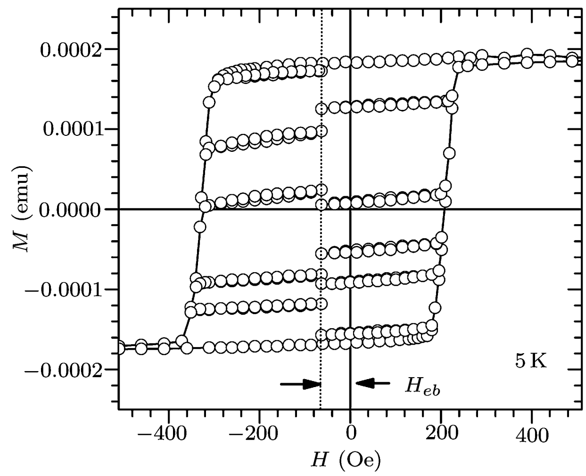

Fig. 2. Recoil loops measured at 5 K after field cooling in 1.5 kOe.

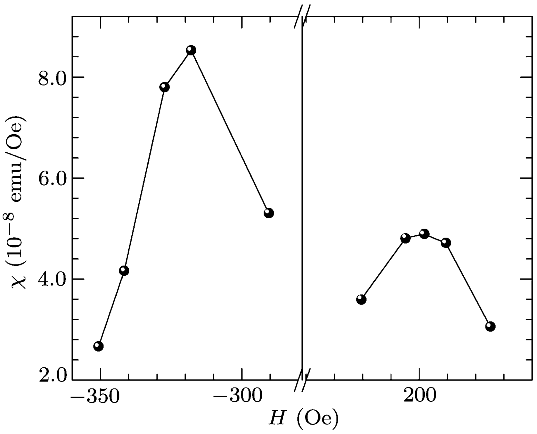

Fig. 3. Recoil susceptibility vs demagnetizing fields calculated from the recoil loops shown in Fig. 2.

Based on a mean field approach, Al-Rsheed and Ei-Hilo have calculated the recoil loops for the granular systems which have aligned easy axes and a distribution of orientation of easy axes, respectively.[24] Their numerical modelling indicates that, regardless of whether the system is completely irreversible (aligned) or it contains both reversible (soft magnetic phase) and irreversible components (hard magnetic phase) of magnetization, the opened recoil loops can be predicted if there is a local interaction field arising from the inter-particle coupling that is strong enough to cause irreversible changes of particles' moments during the recoil process. For the current AF–FM bilayer system, we ascribe the origin of the local interaction field to the interfacial AF–FM exchange coupling. The marked eye effect *only* emerging in the second quadrant of the major loop indicates that the exchange anisotropy in a unidirectional rather than biaxial or uniaxial nature plays a crucial role in the asymmetric magnetization reversal.

On the other hand, in our case the measuring field is parallel to the cooling field which is along one of the biaxial easy axis of LSMO layer. If the unidirectional anisotropy (K_b) is *exactly along* the field direction, we may write the free energy density of the system as $E = -HM\cos\theta + K_b\cos\theta + K_C\cos^2\theta\sin^2\theta + K_{bc}\cos^2\theta\sin^2\theta + K_{bu}\cos^2\theta$, where θ is the angle between field and magnetization vector, the first term is the Zeeman energy, the third term represents the cubic crystalline biaxial anisotropy, and the last two terms represent the possible field induced biaxial and uniaxial exchange anisotropies, respectively. As indicated by the initial Meiklejohn–Bean model[1] and the recent study by Camarero *et al.*,[15] the above formulism does not necessarily yield an asymmetric loop shape except for an enhanced loop width (coercivity) and a horizontal loop shift by an amount of $H_{eb} = K_b/M$. It implies that the unidirectional anisotropy or the AF easy axis shall more or less deviate from the field direction [110]. This is reasonable since in LCMO a nonlinear AF structure with spins pointing close to [100] and [010] directions may exist due to the complex exchange interactions between the mixed valence manganese ions.[29] Because a unidirectional anisotropy locates somewhere between the easy axes [110] and [−110], the magnetization reversal in the descending branch would readily occur by an incoherent rotation or an incoherent multi-domain process, whereas in the ascending branch the reversal proceeds via a coherent domain wall nucleation and motion process. Also, due to this distribution of the magnetic easy axes, lowering the demagnetizing field allows the magnetization vector to easily return to the local energy minimum. This contributes appreciable reversible magnetization. Thus, the recoil susceptibility in the descending branch is higher than that in the ascending branch.

We note that in the present work the measurements of recoil loops only treats a specific asymmetry of magnetization reversal. We thus expect that more feature work could be inspired to study the issue of asymmetric magnetization reversal by applying this minor loop approach to other various asymmetric hysteresis loops, for example, the case in which the AF–FM bilayer film is grown with an *in situ* magnetic field and the measuring field is applied with an angle to the cooling field direction.

References

[1] Meiklejohn W H and Bean C P 1956 *Phys. Rev.* **102** 1413
[2] Nogués J and Schuller Ivan K 1999 *J. Magn. Magn. Mater.* **192** 203
[3] Berkowitz A E and Takano K 1999 *J. Magn. Magn. Mater.* **200** 552
[4] Nikitenko V I, Gornakov V S, Dedukh L M, Kabanov Yu P, Khapikov A F, Shapiro A J, Shull R D, Chaiken A and Michel R P 1998 *Phys. Rev.* B **57** R8111

[5] Fitzsimmons M R, Yashar P, Leighton C, Schuller Ivan K, Nogués J, Majkrzak C F and Dura J A 2000 *Phys. Rev. Lett.* **84** 3986

[6] Leighton C, Song M, Nogués J, Cyrille M C and Schuller Ivan K 2000 *J. Appl. Phys.* **88** 344

[7] Leighton C and Schuller Ivan K 2001 *Phys. Rev.* B **63** 174419

[8] Panagiotopoulos I, Moutis N and Christides C 2002 *Phys. Rev.* B **65** 132407

[9] Gierlings M, Prandolini M J, Fritzsche H, Gruyters M and Riegel D 2002 *Phys. Rev.* B **65** 092407

[10] Krivorotov I N, Leighton C, Nogués J, Schuller Ivan K and Dan Dahlberg E 2002 *Phys. Rev.* B **65** 100402(R)

[11] Spenato D, Pogossian S P and Le Gall H 2003 *J. Magn. Magn. Mater.* **262** 294

[12] Beckmann B, Nowak U and Usade K D 2003 *Phys. Rev. Lett.* **91** 187201

[13] Blomqvist P, Krishnan K M and Ohldag H 2005 *Phys. Rev. Lett.* **94** 107203

[14] Ryu K S, Kim D H, Shin S C and Akinaga H 2005 *Phys. Rev.* B **71** 155308

[15] Camarero J, Sort J, Hoffmann A, García-Martín J M, Dieny B, Miranda R, and Nogués J 2005 *Phys. Rev. Lett.* **95** 057204

[16] Paul A, Kentzinger E, Rücker U and Brückel T 2006 *Phys. Rev.* B **73** 092410

[17] Tillmanns A, Oertker S, Beschoten B, Güntherodt G, Leighton C, Schuller Ivan K, and Nogués J 2006 *Appl. Phys. Lett.* **89** 202512

[18] Ji X, Pakhomov A B and Krishnan K M 2007 *J. Appl. Phys.* **101** 09E507

[19] Liedke M O, Liedke B, Keller A, Hillebrands B, Mücklich A, Facsko S and Fassbender J 2007 *Phys. Rev.* B **75** 220407(R)

[20] Harland C, Lewis L H, Chen Z and Ma B M 2004 *J. Magn. Magn. Mater.* **271** 53

[21] Feutrill E H, McCormick P G and Street R 1996 *J. Phys.* D **29** 2320

[22] Goll D, Seeger M and Kronmüller H 1998 *J. Magn. Magn. Mater.* **185** 49

[23] Emura M, Cornejo D R and Missella F P 2000 *J. Appl. Phys.* **87** 1387

[24] Al-Rsheed A and El-Hilo M 2000 *J. Phys.* D **33** 840

[25] Zhang S Y, Zhang H W, Shen B G, Zhao T, de Boer F R and Buschow K H J 2000 *J. Appl. Phys.* **87** 1410

[26] Kang K, Lewis L H, Jiang J S and Bader S D 2005 *J. Appl. Phys.* **98** 113906

[27] Wang Z H, Cristiani G, Habermeier H -U and Bland J A C 2005 *Phys. Rev.* B **72** 054407

[28] Wang Z H, Cristiani G and Habermeier H -U 2003 *Appl. Phys. Lett.* **82** 3731

[29] Fernádez-Díaz M T, Martínez J L, Alonso J M and Herrero E 1999 *Phys. Rev.* B **59** 1277