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Magnetic exchange coupling in bilayers of two epitaxial ferromagnetic oxides

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ABSTRACT

Despite a decade of research effort on $La_{0.7}Sr_{0.3}MnO_3/SrRuO_3$ (LSMO/SRO) bilayers (BLs), a full knowledge on the magnetic properties and integration of these BLs on silicon substrate is not yet in sight. In this paper, we report on the magnetic exchange coupling observed from the above two ferromagnetic oxide thin film BLs, prepared through a novel approach, called 'domain matching epitaxy'. LSMO (100 nm)/SRO (45 nm) and LSMO (31 nm)/SRO (45 nm) bilayers have been epitaxially integrated with Si (100). Notably, in the former sample, positive exchange bias is observed – an indication of antiferromagnetic exchange coupling and is found to be absent in the latter. Furthermore, in the former sample, the cross-over from antiferromagnetic to ferromagnetic interface exchange coupling is noticed by varying the cooling field. We have verified that this coupling is of magnetic origin, not due to electrostatic interaction by inserting a thin (~10 nm) $SrTiO_3$ layer between LSMO and SRO. We believe that in addition to the formation of interface domain walls, the strong interplay among Zeeman, anisotropic and exchange energies could play a dominant role. Our results would have important implications for devices comprising of magnetic exchange coupled systems.

Magnetic oxide heterostructures of transition metals [1,2] offer a unique opportunity to tailor and tune the magnetic interactions by exploiting local symmetry breaking, epitaxial strain, frustration or charge transfer between the material layers giving rise to very unexpected emergent phenomenon. One of the more frequently studied systems has been $La_{0.7}Sr_{0.3}MnO_3/SrRuO_3$ (LSMO/SRO) bilayers (BLs). For instance, almost a decade ago, Ke and co-authors [3,4] reported on the magnetic properties of this BLs. Although, LSMO/SRO has been envisaged to have tremendous utility in magnetic tunnel junction and magnetic access memory devices, the earlier works did not probe the magnetic properties of this bilayer when the thickness of the individual layer is varied. In addition, to our knowledge, all the previous studies made on LSMO/SRO BLs have only been reported when they were deposited on the closely-lattice matched *non-silicon* substrates such as $SrTiO_3$ (STO) and MgO, which are not compatible with the ubiquitous microfabrication technologies based on the silicon platform and would ultimately limit the application of these materials for magnetic memory devices. The work presented here aims to address the above issues.

Si (100) is the most desirable platform for the development of multifunctional oxide-based nanoelectronic devices, including field-effect transistors and non-volatile memory elements. LSMO is a conducting double exchange ferromagnetic (FM) metal at room temperature. LSMO is frequently used as a active switching layer due to its weak anisotropy and small coercive field (H_c) (~200 Oe). It has a Curie temperature (T_C) of about 360 K. SRO [3,4] is an itinerant FM that makes for a good biasing layer due to its large anisotropy and large H_c (~5000–6000 Oe). The T_C of SRO is near 160 K. SRO is also a commonly used electrode material for ferroelectric oxides. Both oxides have high chemical stability, good thermal properties, and a shared perovskite crystal structure that makes for an easy integration with related oxide materials to form heterostructures.

In this paper, we have reported on our observations of positive exchange bias and cross-over from antiferromagnetic coupling to ferromagnetic coupling in one of the two LSMO/SRO BLs studied, when they were deposited on Si (100). After successfully reproducing the characteristic features of these BLs on silicon wafers, we discovered that these features are strongly dependent on the thickness of biasing (LSMO) layer, which is the most significant finding of this work. The reported observation led us to believe that these anomalous magnetic properties might result from the strong interplay of magneto crystalline anisotropy,

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Zeeman and exchange energies, in addition to the formation of interface domain walls.

Pulsed laser deposition (PLD) was used to grow the epitaxial layers on Si (100) substrates in this work. We have grown $La_{0.7}Sr_{0.3}MnO_3/SrRuO_3/SrTiO_3$(STO)/MgO/TiN/Si (100) heterostructures and optimized the growth conditions for each layer in this geometry. This work is facilitated by the deposition of an epitaxial TiN (a = 4.24 Å) layer, in which, three lattice plans of Si (a = 5.43 Å) match very well with four of TiN (a = 4.24 Å) and the epitaxial growth occurs via domain matching epitaxy (DME) [5–8]. Each layer in this heterostructure was grown epitaxially. This buffer layer consists of TiN, MgO (a = 4.22 Å), and STO (a = 3.905 Å). TiN was chosen because it grows epitaxially on Si(100) and has superior diffusion barrier properties. TiN has an excellent lattice match with MgO, which has a misfit of about 8% with STO. Finally the lattice constant of STO (3.905 Å) matches closely with that of LSMO (3.85 Å) and SRO (3.923 Å). This selection of buffer layers made it possible to integrate epitaxial thin film of LSMO/SRO on Si(100). The prototype bilayers based on these oxides were thoroughly characterized with in-plane and out-of-plane XRD, TEM and STEM-Z techniques as reported [5] in our earlier work. The temperature- and magnetic-field dependent magnetization measurements were carried out using a Quantum design MPMS SQUID dc magnetometer with the sensitivity ⩽10–8 emu at 0 T. The magnetic field is applied along [100] direction of the sample.

In Fig. 1, we present the isothermal magnetic characteristics of two BL structures, namely, sample F: LSMO(100 nm)/SRO(45 nm) and sample G: LSMO(31 nm)/SRO(45 nm). It should be noted that the sample F in this study is the same as sample B in our previous work [5], where, we investigated the properties of this BL by varying (180–45 nm) the SRO layer thickness, keeping LSMO thickness constant at 100 nm. Here, the SRO thickness was kept constant (at 45 nm) as are the thicknesses of the other non-magnetic buffer layers STO (260 nm), MgO (60 nm), and TiN (100 nm). Measurements were performed by cooling each sample under the application of +4T (biasing field) from 400 K down to 4 K to saturate both layers. In each measurement, the sequence of magnetic field sweep is: 0 → +1T → 0 → −1T → 0 → 1T. As one can see, both samples show distinct magnetic reversal features. The most important observations are: (a) pronounced M–H loop shift in sample F toward positive field axis, called positive exchange bias [3,4] (PEB). Interestingly, that shift is found to be absent in sample G; and the appearance of strong high field hysteresis in sample G is absent in sample F. Sample F is showing exchange-spring like behaviour, where the switching behaviour of the soft layer (LSMO) is fully reversible with fields below the transition field of the hard layer (SRO) [9]. Though, we observed switching of hard layer (SRO) at around 5000 Oe in sample G, no such switching is noticed in the SRO layer of sample F, i.e. the SRO moment is frozen. Single layer LSMO and SRO films on Si (100) were also grown as references. Their magnetic properties such as switching characteristics, symmetric hysteresis loops are as one would expect (data not shown), resembling those of pristine bulk samples.

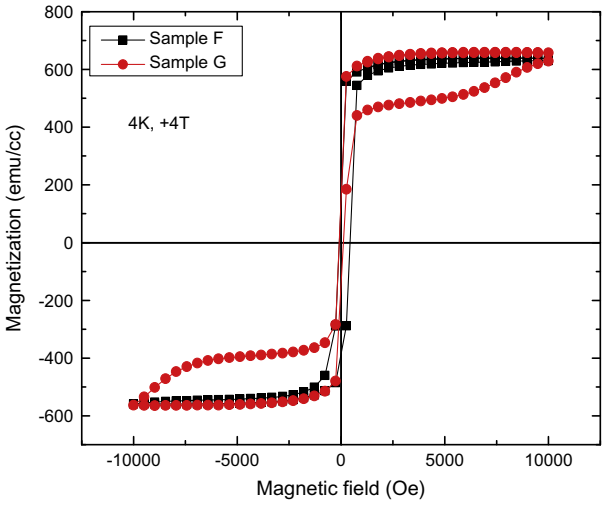

Fig. 1. Comparison of isothermal (4 K) magnetization reversal processes of sample F and sample G. This data were resulted after the samples were field cooled under +4T. Here, we kept the SRO layer thickness constant at 45 nm. As it can be seen, the magnetization reversal processes are distinctly different. Another notable observation is that a positive exchange bias (shift of hysteresis loop toward field cooled direction) is observed only for the former sample.

We focus our attention on sample F which is unique as it shows interesting positive exchange bias (PEB) and exchange-spring behaviour, worth exploring further. To gain additional insights, in Fig. 2(a and b), we further demonstrate and establish the presence of PEB at 4 K on sample F when it is cooled under negative and positive magnetic (biasing) fields (0.2–4T). We used two magnetic field sweeping sequences: (a) 0 → +1T → 0 → −1T → 0 → 1T (full loops of SRO and LSMO) and (b) 0 → +1500 Oe → 0 → −1500 Oe → 0 → 1500 Oe (full loops of LSMO and minor loops of SRO) (see, Fig. S3) [10]. In both the full and minor loop cases, we obtained identical results i.e. M–H loop shifts toward cooling field (CF) direction. When the sample is negative field cooled, the M-H loop shifted toward negative field axis and vice versa. In the previous studies [3,4], this behaviour has been attributed to antiferromagnetic (AFM) coupling at the LSMO/SRO interface. We see no PEB (see Fig. S1 in Supplementary information) [10] above the T_C of SRO implying that the PEB is of magnetic origin. In addition, we have observed no training effect on this sample up to about 5 cycles and see no change in exchange bias, thus ruling out training effect's contribution toward PEB (see Fig. S2 in Supplementary information) [10]. Also, we have verified that the coupling between the layers is not magnetostatic by inserting a thin (∼10 nm) non-magnetic STO insulator layer between SRO and LSMO (data not shown). The magnetization of this modified structure shows no shift at 4 K (no PEB), thus demonstrating that the magnetostatic couplings are small, and the loop shift arises from the magnetic interactions at the SRO/LSMO interface.

The correlations between PEB field (H_{EB}) and coercive field (H_C) may provide important clues as to the microscopic origin of exchange coupling. In Fig. 3(a and b), we plot the cooling field (CF) and temperature dependence of H_{EB} (on left axis shown in black) and H_c (on right axis shown in blue) for sample F under negative field cooling. The results were reproduced when the sample was positive field cooled (see Fig. S3 (a) (b) in Supplementary information) [10].When the cooling field increased above 0.01 T, the H_{EB} changes its sign from positive to negative and H_{EB} saturates as the CF increased further up to 0.9 T. In addition, H_C is the largest in samples showing zero H_{EB}. Such change of sign in H_{EB} has been observed in earlier studies reported on LSMO/SRO BLs [3,4], and in superlattices (SLs) of LSMO/SRO, LSMO/$La_{0.7}Sr_{0.3}CoO_3$ and $Pr_{0.7}Ca_{0.3}MnO_3$/SRO deposited on non-silicon STO substrates [4,5,11,12]. It was explained in terms of formation of Bloch-like interface domain walls and charge redistribution at the manganite/SRO interface. In addition to the shift along the magnetic field axis, we noticed there was also a shift along the magnetization axis when the sample was field cooled. The variation of SRO (derived from the shift along the magnetization axis) magnetic moment as a function of CF is shown in the Supplementary information [10] (see Fig. S4) (discussed later). As shown in the Fig. 3(b), under the CF of 0.1 T, H_C continues to increase as the temperature drops, whereas H_{EB} saturates shortly after the temperature is lowered below the T_C of SRO. The temperature dependence of H_{EB} and H_C are dissimilar, which could be due to the fact that H_{EB} and H_C are