Supporting Information

Ultrathin scale tailoring of anisotropic magnetic coupling and anomalous magneto-resistance in $SrRuO_3 - PrMnO_3$ superlattices

Antarjami Sahoo,[1] Wilfrid Prellier,[2] and Prahallad Padhan[1*]

[1]*Department of Physics, Indian Institute of Technology Madras, Chennai 600036, India*

[2]*Laboratoire CRISMAT, CNRS UMR 6508, ENSICAEN, 6 Bd du Marechal Juin, F-14050 Caen Cedex, France*

AUTHOR INFORMATION :

Corresponding Author :

Padhan@iitm.ac.in

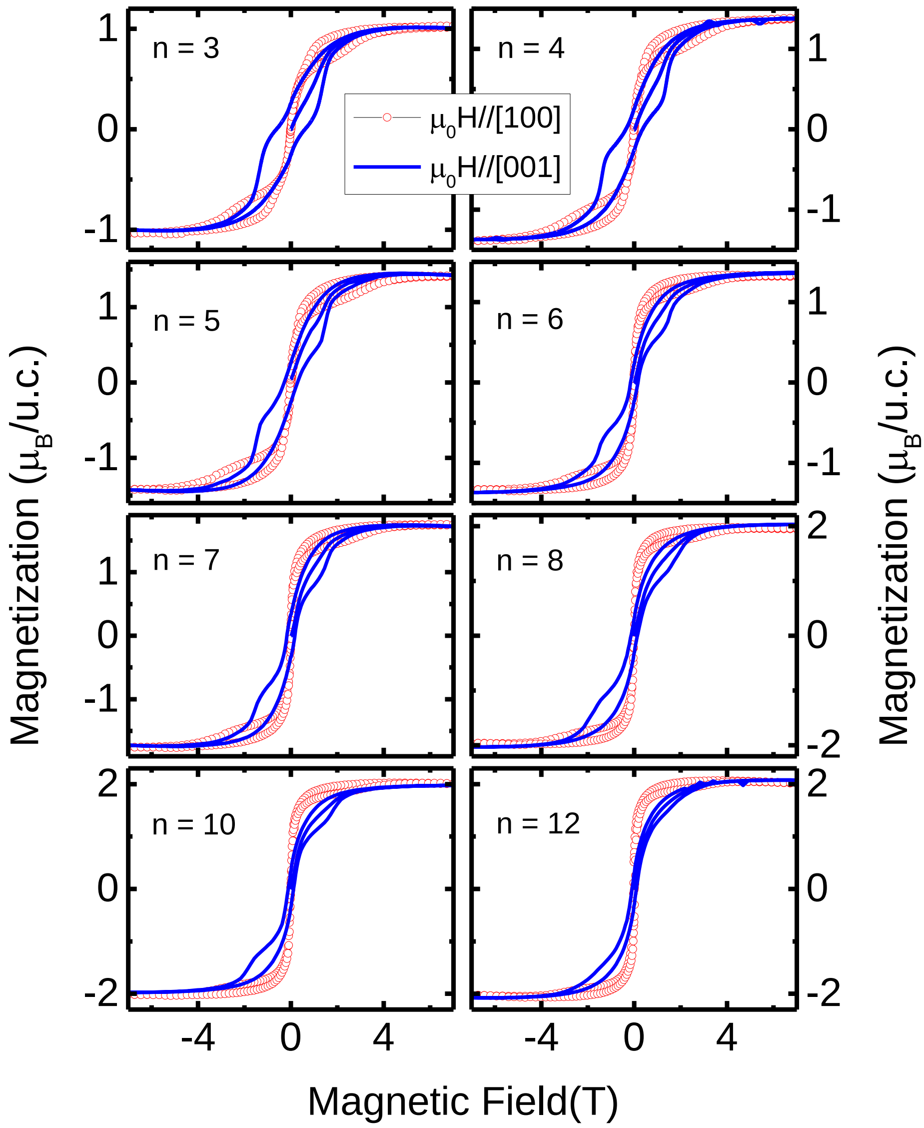

Figure S1: Zero-Field-Cooled field dependent in–plane and out-of-plane magnetization of $(001)STO/[17\,u.c.\,SrRuO_3/n\,u.c.\,PrMnO_3]_{15}$ superlattices measured at $20\,K$.

The zero-field-cooled field dependent magnetization of various superlattices with different PMO space layer measured at $20\,K$ are shown in Figure S1. The magnetization was measured for the field oriented along $[100]_{PC}$ and $[001]_{PC}$ directions. All the superlattices exhibit double hysteresis loop.

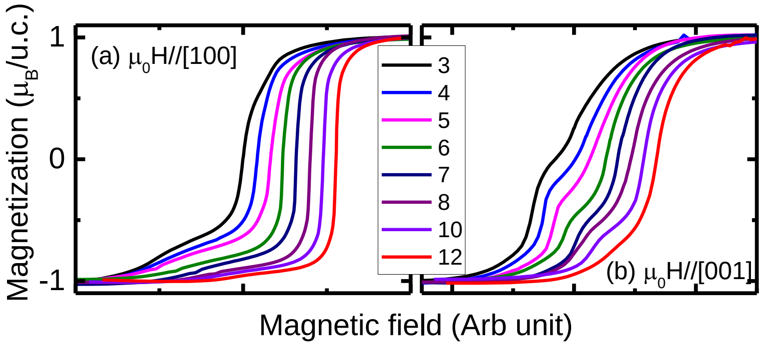

Figure S2: Normalized (a) in-plane and (b) out-of-plane ZFC magnetization of the field decreasing branches of the hysteresis loops of various superlattices. The field decreasing branch of each superlattice is shifted along the field axis.

The in-plane and out-of-plane magnetizations shown in Figure S2 are normalized with magnetic moment at highest applied field to get a qualitative insight of the size of the biased layer at the interfaces. The normalized magnetization of the field decreasing branches of the $M(H)s$ for $[100]_{PC}$ and $[001]_{PC}$ orientations of magnetic field are plotted in Figure S2(a) and S2(b), respectively. The origin is shifted along the field axis by $0.4\,T$ and labelled with the arbitrary unit. The loop opening, which is proportional to the biased SRO layer thickness systematically decreases with the increase of PMO spacer layer thickness.

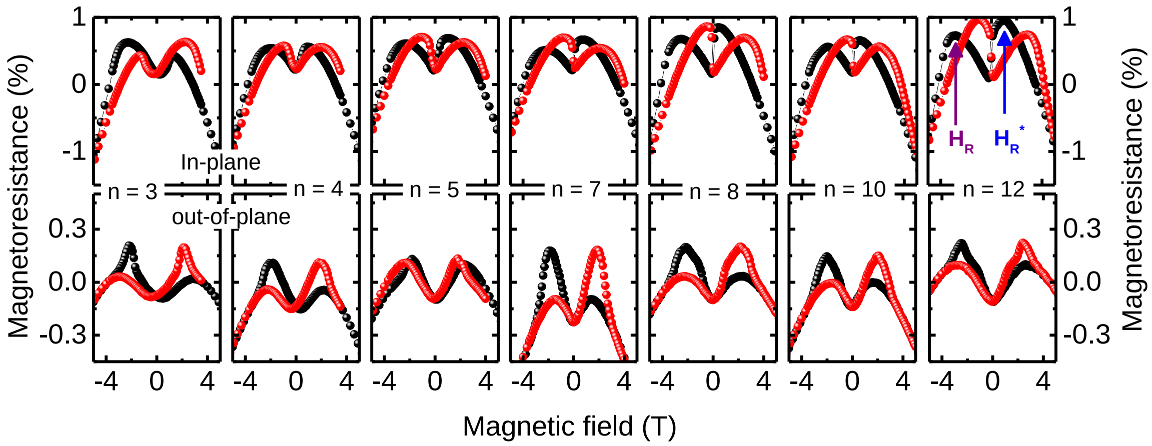

Figure S3: Zero-Field-Cooled field dependent in–plane and out-of-plane magnetoresistance at 20 K of (001)$STO/[17\ u.c.SrRuO_3/n\ u.c.PrMnO_3]_{15}$ superlattices.

Figure S3 shows the zero-field-cooled (ZFC) field dependent magnetoresistance $\left(MR(H) = \frac{R(H)-R(0)}{R(0)}\right)$ measured at 20 K for various superlattices with field oriented along $[100]_{PC}$ and $[001]_{PC}$ directions. The MR is negative at 7 T field owing to the spin polarised d-band of SRO (see Figure 4(c) and 4(d)). As the field decreases below 7 T, the angular separation of the spins of pinned SRO near the interfaces and free SRO (see Figure S4) induce additional spin dependent scattering. Thus, it decreases the probability of transport of Ru 4d electrons from one free SRO to another, which leads to a larger resistance compare to the zero-field resistance, i.e. the MR changes its sign and becomes positive. On further decreasing the field, the spins of the pinned SRO layer, influenced by the rotation of Mn spins because of their exchange coupling nature, start rotating toward the free layer at a field H_R^*. This decreases the resistance and hence, the MR. The decrease of in-plane MR of the superlattices with $n =$ 3 to 6 is steady due to the gradual rotation of spins. While the variation of in-plane MR of the superlattices with $n \geq 7$ is faster due to easy spin flipping of biased SRO. Once the Mn spins flip, and the free SRO rotates, the pinned SRO start inducing the spin dependent scattering and the resistance increases up to the field H_R. The anomalous behaviour of MR

between H_R^* and H_R of the superlattices is mainly determined by (i) electrical conduction of Ru $4d$ electrons in SRO and (ii) the probability of transport of Ru $4d$ electrons from one SRO layer to another through the PMO layer in the presence of magnetic field. The H_R corresponds to the H_C of the superlattices, a field at which the Zeeman energy overcomes the exchange coupling energy and the biased Ru spins start rotating towards the field direction.

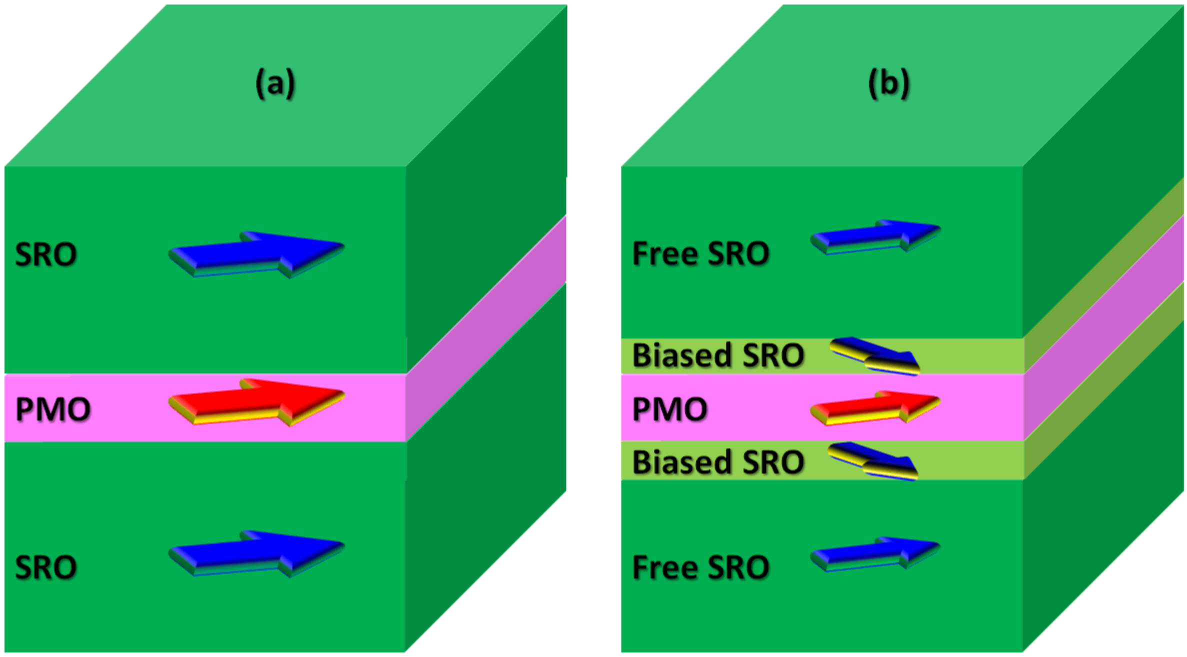

Figure S4 : Schematics of (a) an ideal and (b) an as prepared $SRO - PMO$ superlattice, where the arrows indicate the orientation of spins of different layers, created by the in-plane applied field larger than the coercive field of thin film of SRO.

As the SRO and PMO are ferromagnetic, the net spin orientations of each layer in the superlattice for the ideal case (i.e. in the absence of exchange coupling between the layers) is expected to be along the direction of the field (see Figure S4(a)). However, in presence of strong exchange coupling i.e. in the present case, the strong AFM coupling at the interfaces, the SRO can be visualized as a combination of the interfacial biased part (pinned) and unbiased part (free) (see Figure S4(b).