Supplementary Information:

Anharmonic phonon interactions and Kondo effect in FeSe/Sb₂Te₃/FeSe heterostructure: Proximity effect between ferromagnetic chalcogenide and dichalcogenide

Labanya Ghosh¹, Mohd Alam¹, Mahima Singh¹, Srishti Dixit¹, Satya Vijay Kumar¹, Abhineet Verma², Prashant Shahi³, Yoshiya Uwatoko⁴, Satyen Saha², Archana Tiwari⁵, Ajay Tripathi⁶ and Sandip Chatterjee^{1,*}

¹Department of Physics, Indian Institute of Technology (BHU) Varanasi 221005, India ²Department of Chemistry, Banaras Hindu University, Varanasi 221005, India ³Department of Physics, D.D.U. Gorakhpur University, Gorakhpur 273009, India ⁴Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 2778581, Japan ⁵Department of Physics, Institute of Science, Banaras Hindu University, Varanasi 221005, India ⁶Department of Physics, School of Physical Sciences, Sikkim University, Gangtok, Sikkim 737102,

India

*Corresponding author: <u>schatterji.app@iitbhu.ac.in</u>

SI1. Structural confirmation

We have discussed the structural, morphological, physical and magnetic properties of FeSe/Sb₂Te₃/FeSe hetero-structure in our original manuscript. the thickness of the hetero-structure was around ~75.3 nm, the grazing incidence (GI) angle was fixed at 0.2° . At such a low GI angle, the penetration depth remains exceptionally shallow, and thus the penetrated X-rays propagate almost parallel to the film surface and come out by diffraction from the lattice planes. In this way, an out-of-plane X-Ray Diffraction (XRD) scan provides structural information on thin films by nullifying the substrate effect. Thus, sometimes the diffraction peak signal becomes very weak compared to the background signal and appears to be unclear due to low thickness. For our case, the FeSe layer thickness is much less (~15 nm) as compared to the Sb₂Te₃ layer (~50 nm). So, the peaks corresponding to FeSe did not appear well in the diffraction pattern. Thus, to confirm the structural information, we studied the diffraction pattern for the hetero-structures of larger thicknesses and compared them with the diffraction peaks corresponding to Sb₂Te₃ and FeSe targets as shown in the Fig. S1. The diffraction peaks are more significant for higher thicknesses as expected. For the 428.8 nm thick hetero-

structure all the peak positions corresponding to FeSe and Sb_2Te_3 are visible as shown in the inset of Fig. S1.



Fig. S1: The X-Ray Diffraction patterns of as-prepared hetero-structures compared with Sb₂Te₃ single crystal and FeSe polycrystal. Inset shows a closer view of the 428.8 nm thick hetero-structure.

SI2. Comparing magnetic properties of as-prepared hetero-structures

We measured the magnetic moments for all the prepared hetero-structures of different thicknesses concerning temperature from 300 to 700 K. The value of magnetic moment differs with varying thicknesses as shown in Fig. S2. All samples show usual magnetization with temperature. However, it was surprising to observe that, the hetero-structure with 75.3 nm thickness attributes an unusual magnetic anomaly in the temperature range 465 K to 485 K in both ZFC/FC magnetization curves, which is demonstrated as a shaded region in the inset of Fig. 5(d) in the original manuscript and compared with other hetero-structure in Fig. S2 here. This dip in magnetization was described as a certain local spin ordering in the vicinity of the interface of FeSe and Sb₂Te₃ that appeared due to lattice distortion and phonon vibrations at high temperatures. Besides this, the charge order driven local spin ordering might also be a reason behind such anomaly in M-T curves in the vicinity of 475 K (T_{sp}) temperature as mentioned in the original manuscript. Thus, it can be understood unambiguously that there is some short-range local magnetic ordering in the 75.3 nm system around 475 K.

But increasing the thickness, the up written drop disappeared indicating that there is no local ordering in those systems. The enhancement of thickness directly corresponds to the increasing bulk behavior in the system. For 91.9 nm, 271.8 nm, and 428.8 nm hetero-structures, the intrinsic bulk effect dominates over the interfacial proximity effect diminishing the probability of proximity induced local spin ordering. Further, in the 75.3 nm film, spin-phonon coupling initiated by local charge ordering at the proximity of the interface in the prepared hetero-structure was confirmed using the Raman spectroscopy study. We measured moment vs. magnetic field for all the heterostructures at 350 K and 580 K, which signifies the similar ferromagnetic behavior as shown in Fig. S3 (a-d). The change in saturation magnetization is larger for the 75.3 nm thick sample as compared to others might be due to the presence of local spin ordering in-between. Thus, we have measured the magnetic properties of the heterostructures with different thicknesses. But we observed local magnetic ordering only in the 75.3 nm hetero-structure, so we measured the Raman spectra and transport properties only for this system to understand the phenomena behind such behavior. After all observations, we concluded that due to the dominancy of the interfacial states in the 75.3 nm system, local spin evolution was dominated over the intrinsic bulk magnetization, and this local spin evolution initiated a short-range ordering as shown in Fig. S2.



Fig. S2: The magnetic moment vs. temperature for the hetero-structures of different thicknesses.



Fig. S3: Magnetic moment variation with magnetic field for the hetero-structures of thicknesses (a) 428.8 nm, (b) 271.8 nm, (c) 75.3 nm, (d) 91.9 nm.