

Supplementary Information

Heterostructured Ferromagnet-Topological Insulator with Dual-phase Magnetic Properties

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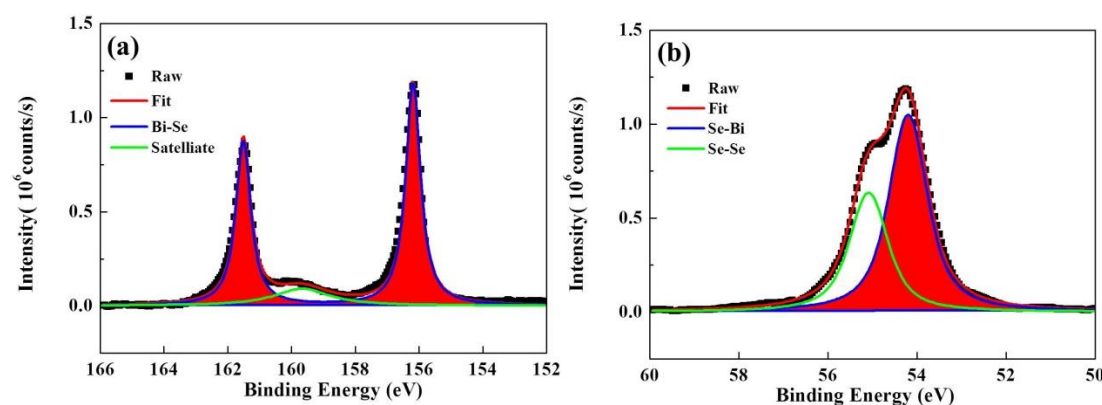


Fig. S-1 SR-PES spectra of (a) Bi-4*f* and (b) Se-3*d* where highlighted areas are used for Bi/Se ratio calculation, with the use of following equation:

$$n = \frac{I}{S}$$

where

$$S = f\sigma\theta y\lambda AT$$

where f is the Photon flux of the x-ray, σ is the photoionization cross-section of peak of element, θ is the photoelectron emission angle, y is the photoelectron yield, λ is the inelastic electron mean-free path, A is analysis area and T is a analyzer transmission function. Here, we define S is the sensitivity factor of core level of element.

Hence, n is dependent on the peak area and sensitivity factor.

$$\frac{n_1}{n_2} = \frac{(I_1/S_1)}{(I_2/S_2)} \quad (1)$$

The calculated Bi/Se ratio is 1.49 very close to the ideal stoichiometry of Bi_2Se_3 formula.

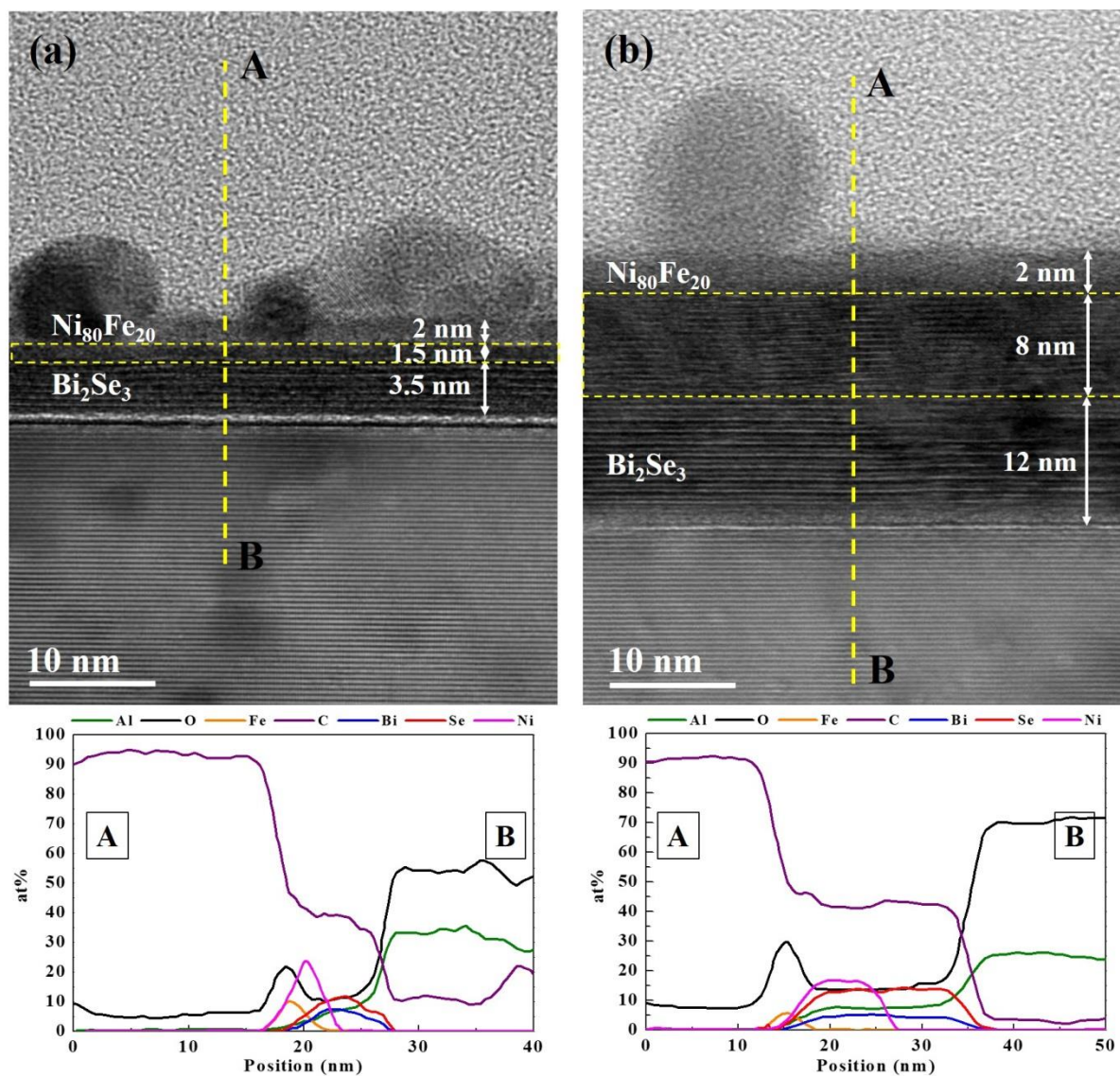


Fig. S-2 Cross-sectional TEM images of (a) $\text{Py}/\text{Bi}_2\text{Se}_3$ -5nm and (b) $\text{Py}/\text{Bi}_2\text{Se}_3$ -20 nm samples. EDX analyses are shown below the corresponding TEM images, where EDX line-scanning was performed along the yellow dashed line which starts from A and ends at B. EDX shows Ni diffusion into Bi_2Se_3 and it formed an Ni-Se overlapping region.

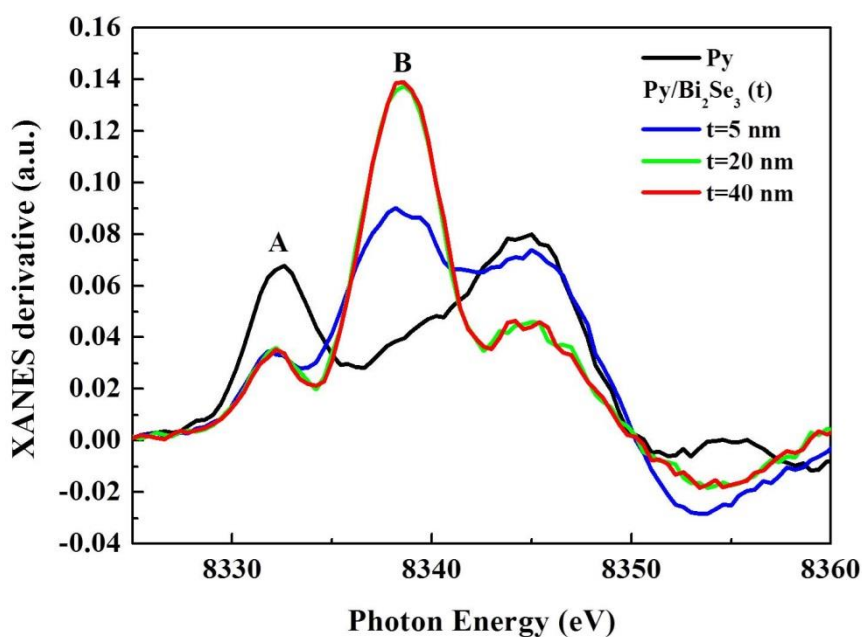


Fig. S-3 Differentiation of Ni *K*-edge XANES for pure Py (black), Py/Bi₂Se₃-5nm, Py/Bi₂Se₃-20nm and Py/Bi₂Se₃-40nm samples. The edge characteristics can be decomposed into feature A and B upon differentiation. Feature A is allowed by quadrupole selection rules. The weak intensity corresponds to a quadrupole $1s \rightarrow 3d$ dipole-forbidden transitions as a characteristic of chemical environment in Ni valency in which the intensity strongly depends upon the coordination symmetry. Feature A becomes less intense in Ni:Bi₂Se₃ as compared to Py, suggesting a local-structural change to centro-symmetry in coordination [1-3]. Feature B is attributed to the $1s \rightarrow 4p$ transition, which arises from a chalcogen-based charge-transfer and indicates the presence of strong Ni-Se covalent bonds. Feature B is intensified with the increase of Bi₂Se₃ thickness, implying a more stabilized phase of Ni:Bi₂Se₃. On the contrary, feature B is heavily suppressed in pure Py, which is a straightforward indicator to the absence of Ni:Bi₂Se₃.

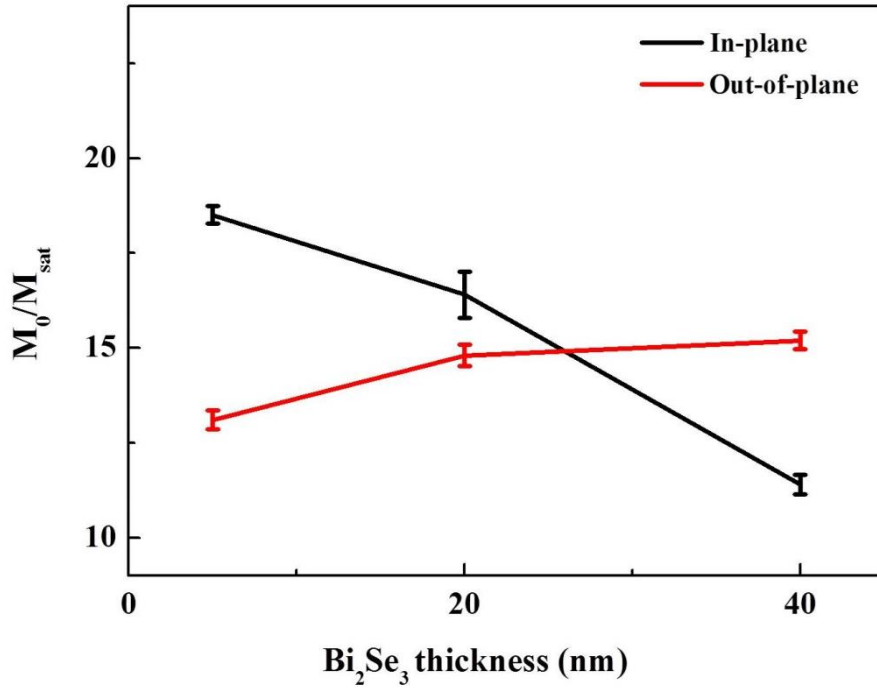


Fig. S-4 The dependence of remanence ratios (M_0/M_{sat}) on Bi_2Se_3 thickness, for in-plane and out-of-plane directions. M_0/M_{sat} was estimated from M-H curves.

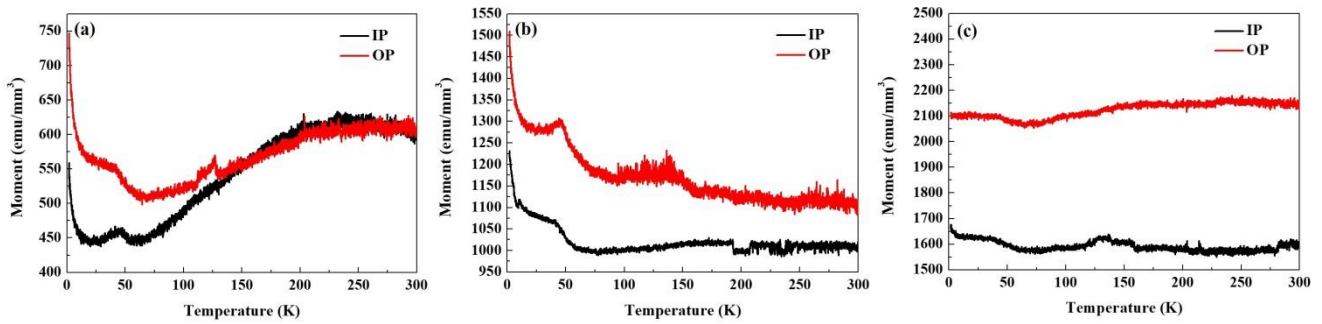


Fig. S-5 The temperature dependence of magnetization of (a) $\text{Py}/\text{Bi}_2\text{Se}_3$ -5 nm, (b) $\text{Py}/\text{Bi}_2\text{Se}_3$ -20 nm, (c) $\text{Py}/\text{Bi}_2\text{Se}_3$ -40 nm, for in-plane (black) and out-of-plane (red) directions.

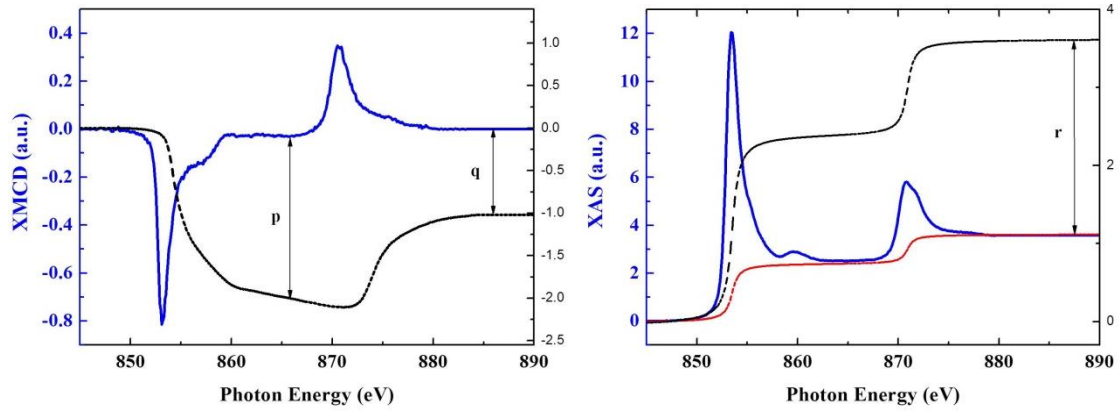


Fig. S-6 (left) Ni-XMCD and (right) Ni-XAS integrations applied in sum-rules analysis for the calculation of Ni's ΔE_{SO} . The orbital (m_{orb}) and spin (m_{spin}) moments are obtained by following equations:

$$m_{orb} = -4q(10 - n_{3d}) / 3r \quad (1)$$

$$m_{spin} = -(6p - 4q)(10 - n_{3d}) / r \quad (2)$$

Where

$$q = \int_{L3+L2} (\mu_+ - \mu_-) d\omega$$

$$r = \int_{L3+L2} (\mu_+ + \mu_-) d\omega$$

$$p = \int_{L3} (\mu_+ - \mu_-) d\omega$$

where μ_+ / μ_- is the x-ray absorption intensity with left/right circular polarization; n_{3d} is the number of 3d electron per cation and the p , q and r are integrated values obtained from XMCD and XAS.

The calculated Ni m_{orb} are 0.02, 0.018 and 0.01 for Py/Bi₂Se₃-5 nm, Py/Bi₂Se₃-20 nm, and Py/Bi₂Se₃-40 nm, respectively, along in-plane direction, which is denoted m_{orb}^{\parallel}

Ni m_{orb} are 0.07, 0.07, 0.06 for Py/Bi₂Se₃-5 nm, Py/Bi₂Se₃-20 nm, and Py/Bi₂Se₃-40 nm, respectively, for out-of-plane direction, which is denoted m_{orb}^{\perp} .

In a simple model of Bruno [4][5], a relation between m_{orb} and ΔE_{SO} is derived as:

$$E_{SO} = -\frac{1}{2} \frac{\xi}{2\mu_B} [m_{orb}^{\perp} + (m_{orb}^{\parallel} - m_{orb}^{\perp}) \sin^2 \theta]$$

The energy difference for the magnetization aligned along out-of-plane and in-plane directions is:

$$\Delta E_{SO} = E_{SO}(\theta = 0^\circ) - E_{SO}(\theta = 90^\circ)$$

$$\Delta E_{SO} = -\frac{1}{2} \frac{\xi}{2\mu_B} [m_{orb}^{\perp} - m_{orb}^{\parallel}]$$

where ξ is spin-orbit coupling parameter for 3d electron and the values of m_{orb}^{\perp} and m_{orb}^{\parallel} are obtained by

sum-rules [6] along the out-of- and in-plane directions, respectively.

The calculated $m_{orb}^{\parallel} - m_{orb}^{\perp}$ values are 0.02795, 0.02907 and 0.02795, for Py/Bi₂Se₃-5 nm, Py/Bi₂Se₃-20 nm, and Py/Bi₂Se₃-40 nm, respectively.

The same method was used in the calculation of Fe's ΔE_{SO} .

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