Supporting information

Ferromagnetic anomalous Hall effect in Cr-doped Bi_2Se_3 thin films via surface-state engineering

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Content

I. The anomalous Hall effect in a thinner film: 1-6-1 QL structure of $Ca_{0.04}(Cr_{0.5}Bi_{0.5})_{1.96}Se_3$ – $Ca_{0.04}Bi_{1.96}Se_3$ – $Ca_{0.04}(Cr_{0.5}Bi_{0.5})_{1.96}Se_3$ films

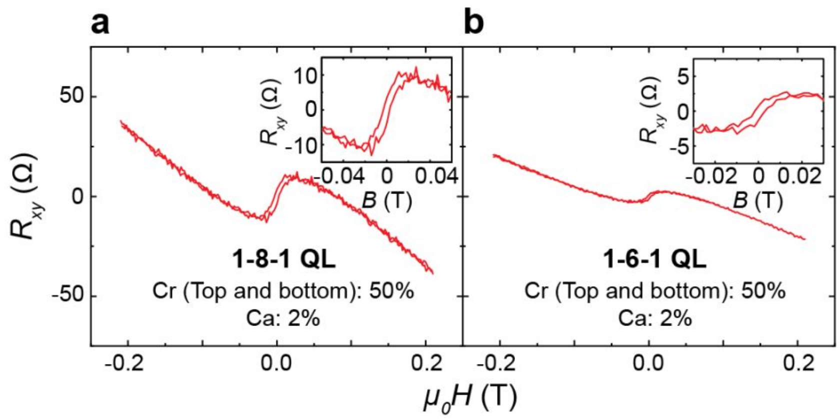

Figure S1. The anomalous Hall effect signals in the surface engineered films. Hysteresis loops observed in (a) 1-8-1 QL and (b) 1-6-1 QL structure of $Ca_{0.04}(Cr_{0.5}Bi_{0.5})_{1.96}Se_3$ – $Ca_{0.04}Bi_{1.96}Se_3$ – $Ca_{0.04}(Cr_{0.5}Bi_{0.5})_{1.96}Se_3$ films.

II. Calculation methods

First-principles calculations are carried out by using VASP[1,2] and wannierized by using the VASP-WANNIER90[3] interface to arrive at a tight-binding description of first-principles quality. The pseudopotential is of the projector-augmented-wave type as implemented in VASP[4,5]. The Perdew-Burke-Ernzerhof approximation for solid (PBEsol)[6] is employed to describe the generalized gradient approximation type of exchange-correlation functional. We choose an energy cutoff of 300 eV and the Brillouin zone is sampled with k-point grids of size $10 \times 10 \times 10$ for pristine Bi_2Se_3 and $4 \times 4 \times 1$ for $\sqrt{3} \times \sqrt{3} \times 1$ hexagonal supercell structures in 3 QLs with Cr partially substituting for Bi. Atomic spin-orbit-coupling is added to Bi p and Se p orbital basis after wannier projection.

III. Details for the mass-gap analysis

In Fig. 4a and b, we find that our computed Dirac cone splittings E_g are well described by the formula

$$E_g = \min | E_{mag} \pm E_0 | , \tag{1}$$

where $E_0 = 19\ \mu\text{eV}$ is the gap induced by the interaction between top and bottom surfaces of an 8 QL thick film, and

$$E_{mag} = \chi_{Bi}\,\Delta_{Bi} + \chi_{Se}\,\Delta_{Se} \tag{2}$$

with $\chi_{Bi} = 0.420$ and $\chi_{Se} = -0.034$ being the mass-gap susceptibilities defined as the ratios of the induced mass gap to the applied Zeeman field. Two topological transitions then occur when $E_g = 0$, with the film as a whole exhibiting Chern numbers of $C = -1$, 0, and +1 for $E_{mag} > E_0$, $|E_{mag}| < E_0$, and $E_{mag} < -E_0$, respectively. The mass-gap susceptibility of Se is, in principle, expressed as $\chi_{Se} = 1/3(\chi_{Se1} + 2\chi_{Se2})$, where Se1 and Se2 refer to the central and the two outer Se atoms, respectively. However, the mass-gap susceptibility of Se1 is negligible ($|\chi_{Se1}| < 0.2\ |\chi_{Se2}|$), so the Zeeman splitting of Se1 is not included in the resulting average.

In Fig. 4a and b, the Chern numbers are obtained from the Hall conductivities calculated by the Kubo formula with the Wannier functions as follows.

$$\sigma_{\mathrm{AH},ij} = e^2\hbar \sum_{n\neq n'} \int \frac{d\boldsymbol{k}}{(2\pi)^d}\left[f(\varepsilon_n(\boldsymbol{k})) - f(\varepsilon_{n'}(\boldsymbol{k}))\right] \times \mathrm{Im}\frac{\langle n,\boldsymbol{k}\,|\,v_i\,|\,n',\boldsymbol{k}\rangle\langle n',\boldsymbol{k}\,|\,v_j\,|\,n,\boldsymbol{k}\rangle}{[\varepsilon_n(\boldsymbol{k}) - \varepsilon_{n'}(\boldsymbol{k})]^2} \tag{3}$$

f is the Fermi distribution function, and $\varepsilon_n(\boldsymbol{k})$ is an energy eigenvalue of the n^{th} band at $\boldsymbol{k}$. v_i and v_j are the velocity operators of i and j cartesian directions. d is the dimensionality, so $d = 2$ due to the 2D nature of the topological surface states. The Berry curvature is sampled in a two-dimensional k-point mesh whose grid density corresponds to 100×100 near the Brillouin zone boundary and $10^5 \times 10^5$ near the Γ point. The Fermi level is set to lie in the direct band gap so that the Chern number would be quantized.

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