Supporting Information

Field Electron Emission of Layered Bi₂Se₃ Nanosheets with Atomic-Thick Sharp Edges

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Part 1. Fabrication routing and supplement characterizations

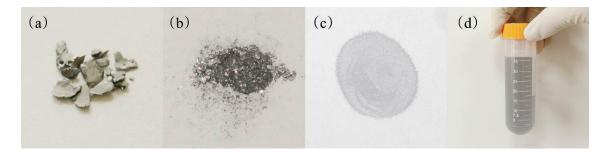


Figure S1. The fabrication routing of layered Bi₂Se₃ nanosheets dispersion. (a) Bulk Bi₂Se₃ crystals. (b) Bi₂Se₃ powders. (c) Layered Bi₂Se₃ nanosheet clusters. (d) Dispersion of layered Bi₂Se₃ nanosheets.

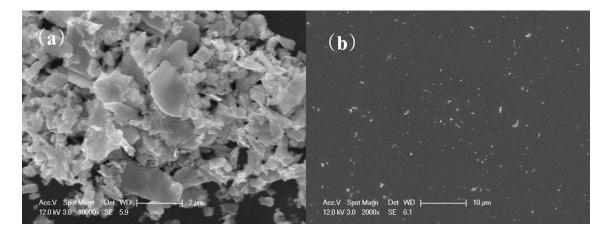


Figure S2. (a) SEM image of the layered Bi₂Se₃ nanosheet clusters. It depicts the SEM image of the hydrothermal exfoliated Bi₂Se₃ nanosheet clusters, which reveals that the hydrothermal intercalation and exfoliation method could turn the Bi₂Se₃ from bulk-size to nano-size (see

optical images from **Figure S1(a) to (c)**). (b) SEM images of the exfoliated layered Bi₂Se₃ nanosheets spin-coated on Si substrate at large scale.

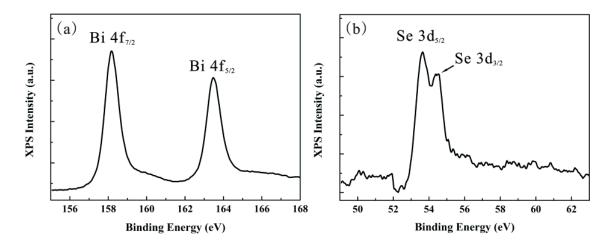


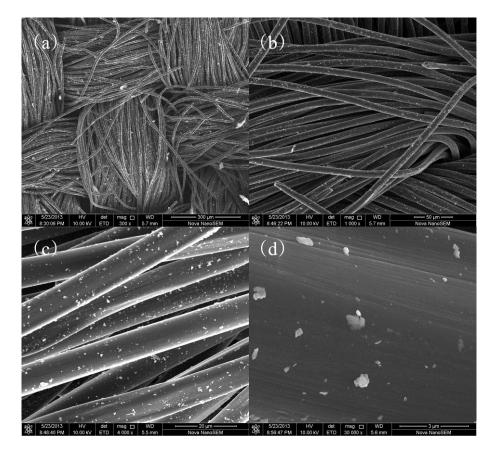
Figure S3. XPS spectra of Bi₂Se₃.(a) bismuth region (Bi4f) and (b) selenium region (Se3d).

Part 2. Field emission of carbon cloth decorated with layered Bi₂Se₃ nanosheets.

The samples were directly fabricated by solution-casted on the pre-clean carbon cloth.



Figure S4 Photograph of carbon cloth decorated without and with layered Bi₂Se₃ nanosheets, which can't be distinguished by naked eyes.



 $\textbf{Figure S5.} \ \text{SEM images of carbon cloth decorated with layered } Bi_2Se_3 \ nanosheets.$

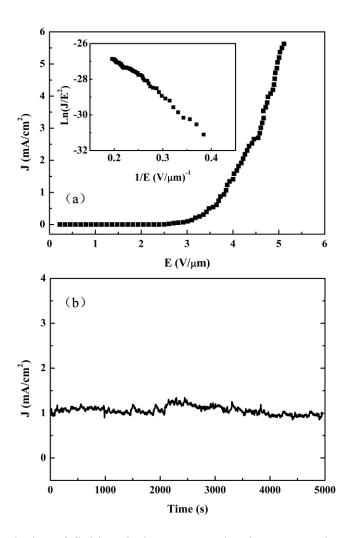


Figure S6 (a) Typical plot of field emission current density versus electric field (J-E) of the layered Bi₂Se₃ nanosheets on carbon cloth. Inset is the corresponding Fowler-Nordheim (FN) plot. (b) The field emission current stability over time.

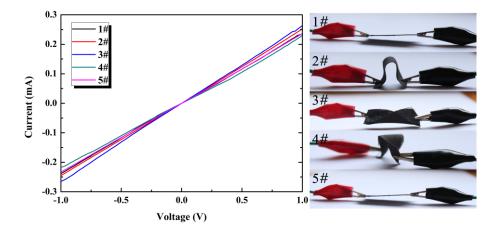


Figure S7. I–V curves of the sample under different bending conditions (1#-4#). 5# records the I–V curve of carbon fabric after 50 cycles of bending, which reveals the high flexibility and stability of the carbon colth with layered Bi₂Se₃ nanosheets on it.

Part 3. Simulation and theoretical calculation

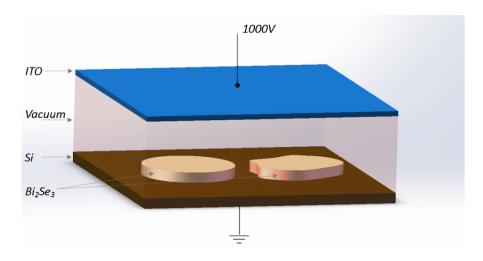


Figure S8. Schematic diagram of simulating electrical filed distribution.

Table S1. Parameters of materials in simulation

	Thickness	Permittivity
ITO	80 nm	7.6
Vacuum	220 μm	1.0
Si	400 μm	11.7
Bi ₂ Se ₃	5 nm	3.7

The permittivity of 5 QL (\sim 5 nm) Bi₂Se₃ was simulated from density functional theory. Our first principles electronic structure calculations were carried out in the projector augmented wave (PAW) framework parametrized by Perdew-Burke-Ernzerhof (PBE) of the generalized gradient approximation (GGA) [1] in the Vienna *ab initio* simulation package (VASP) [2,3]. Spin-orbit interactions (SOC) are turned on to capture the Dirac cone feature of the surface electronic states [4,5]. Fully relativistic PAW pseudo potentials acting on valence electron wave functions were used. The plane-wave kinetic energy cutoff is 400 eV and the self-consistency tolerance is 1×10^{-8} eV. The bulk material structure is relaxed until all force components are less than 0.001 eV/Å. The static dielectric response matrix was determined using density functional perturbation theory with local field effects. The method is explained

- in Ref. [6]. One quintuple layer is defined as in Ref. [4] and the distance between slabs is 10 Å to avoid interlayer interactions. Based on these, the permittivity of 5 QL (\sim 5 nm) Bi₂Se₃ in z direction was calculated as \sim 3.7, which is shown in **Table S1**. More details of which will be shown in our coming work.
- [1] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).
- [2] G. Kresse and J. Hafner, Phys. Rev. B 47, 558 (1993).
- [3] G. Kresse and D. Joubert, Phys. Rev. B 59, 1758 (1999).
- [4] H. Zhang, C. Liu, X. Qi, X. Dai, Z. Fang, and S. Zhang, Nat. Phys. 5, 438 (2009).
- [5] O. V. Yazyev, J. E. Moore, and S. G. Louie, Phys. Rev. B 105, 266806 (2010).
- [6] M. Gajdos, K. Hummer, G. Kresse, J. Furthmuller and F. Bechstedt, Phys. Rev. B 73, 045112 (2006).