

Supplemental Materials for
Evidence of Topological Surface State in Three-Dimensional Dirac
Semimetal Cd₃As₂

Hemian Yi¹, Zhijun Wang¹, Chaoyu Chen¹, Youguo Shi¹, Ya Feng¹, Aiji Liang¹, Zhuojin Xie¹, Shaolong He¹, Junfeng He¹, Yingying Peng¹, Xu Liu¹, Yan Liu¹, Lin Zhao¹, Guodong Liu¹, Xiaoli Dong¹, Jun Zhang¹, M. Nakatake³, M. Arita³, K. Shimada³, H. Namatame³, M. Taniguchi³, Zuyan Xu⁴, Chuangtian Chen⁴, Xi Dai¹, Zhong Fang¹ and X. J. Zhou^{1,2,*}

¹*Beijing National Laboratory for Condensed Matter Physics,
Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

²*Collaborative Innovation Center of Quantum Matter, Beijing, China*

³*Hiroshima Synchrotron Radiation Center,
Hiroshima University, Hiroshima 739-8526, Japan*

⁴*Technical Institute of Physics and Chemistry,
Chinese Academy of Sciences, Beijing 100190, China.*

**Corresponding author: XJZhou@aphy.iphy.ac.cn.*

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1. Growth of Cd₃As₂ Single Crystals.

High quality single crystals of Cd₃As₂ were grown by using vapor phase transition method. The mixtures of As (99.999%) and Cd (99.999%) pieces in stoichiometric ratio were placed in an alumina crucible and sealed in an evacuated quartz tube. The mixture was heated up to 750°C for 20 hours and then cooled down to room temperature. Then the mixture was transferred into the tube furnace and then heated up to 690°C for one week. The evacuated quartz tube was put at a temperature gradient environment and the mixture was put in the high temperature part.

2. ARPES Measurements.

The angle-resolved photoemission measurements were performed on two photoemission systems. For the ARPES measurements on our lab-based system[1] that combines the Scienta R4000 analyzer and a helium discharge lamp (photon energy of 21.218 eV), the overall energy resolution was set at ~ 20 meV and the angular resolution is ~ 0.3 degree. The photon energy-dependent ARPES measurements were carried out at Beamline 7 of the Hiroshima Synchrotron Radiation Center. The beamline is equipped with the Scienta SES2002 analyzer and a He-flow-type five-axes goniometer. The samples were cleaved *in situ* and measured in vacuum with a base pressure better than 5×10^{-11} Torr.

3. Band Structure Calculation Method.

The electronic band structure calculations are performed within the density functional formalism as implemented in VASP. We use the all-electron projector augmented wave (PAW) basis sets with the generalized gradient approximation (GGA) of Perdew, Burke and Ernzerhof (PBE) to the exchange correlation potential[2]. To match the ARPES measurements, we just calculate hypothetical zinc-blende(ZB) structure with a simplest unit of face-center of cubic cell, and modify the energy of s orbital energy from the maximally localized Wannier functions(MLWFs) to take the random effect of the vacancies on the valence bands into account. The spin-orbit coupling was taken into account self-consistently. We construct the MLWFs from the ZB ab initio calculation using the method developed by Marzari and co-workers[3]. The imaginary part of the surface Green's function of the semi-infinite system, obtained by an iterative method, is the local density of states (LDOS), from which we can obtain the dispersion of the surface states. The surface

LDOS on the (112) surface is shown in Fig. 3d, which is consistent with the ARPES data.

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