Supplemental Material for
"Competition between stepwise polarization switching and chirality coupling in ferroelectric GeS nanotubes"

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Methods

AIMD simulations

We optimize the geometry of the unit cell of GeS monolayer and expand it to a 5 × 5 supercell, which consists of 100 atoms (50 Ge atoms and 50 S atoms). The supercell is used as input for the AIMD simulations. We perform AIMD simulations within the canonical (NVT) ensemble using the Langevin thermostat[1,2] and employing Perdew-Burke-Ernzerhof (PBE) functional[3] in the Vienna *Ab initio* Simulation Package (VASP). The projector augmented wave (PAW) potentials are used with a plane-wave energy cutoff of 400 eV[4,5]. Γ-point sampling for the Brillouin zone is adopted. In the AIMD simulations, we set the temperature to 300 K with a timestep of 1 fs. The total energy of the system and forces on each atom are calculated from the AIMD simulations. In order to cover a large sampling space, we prepare eleven systems (supercells with different volumes) to train the deep-learning potential. We run 6000 steps of AIMD for the optimized 5 × 5 supercell, and 1000 steps for the other ten volumes. 15,000 frames of data are used to train the deep-learning potential and the rest employed for validation.

Deep-learning potential

The whole training process of the interatomic potential is carried out using the deep-learning package, DeePMD-kit[6,7]. In the training process, the full relative coordinates of atoms in the supercell are used to construct the descriptors. The number of neighbors of each type of atom is set to 16 and the cut-off radius is 6 Å. The number of neurons in three hidden layers of the embedding network and fitting network are (10, 20, 40) and (120, 120, 120), respectively. The learning rate starts from 1×10^{-3} to 1×10^{-8}. The prefactors of energy and force loss functions at the beginning and limit of the training are used as 0.02, 1, 1000, 1.

Atomistic simulations of GeS nanotubes

We perform atomistic simulations for GeS nanotubes using the newly developed deep-learning potential in the LAMMPS package[8]. The energy minimization and NEB calculations[9,10] using the deep-learning potential are also conducted in LAMMPS. In this work, the indices of the zigzag nanotubes are (200, 0), (250, 0), (300, 0), (350, 0), (400, 0) and the indices of chiral nanotubes are (240, 240), (312, 312), (360, 360), (432, 432), (480, 480). The corresponding supercells contain 800, 1000, 1200, 1400, and 1600 atoms for the zigzag nanotubes, and 2400, 3120, 3600, 4320, and 4800 atoms for the chiral nanotubes, respectively.

References

[1] Hoover W G, Ladd A J and Moran B 1982 *Phys. Rev. Lett.* **48** 1818
[2] Evans D J 1983 *J. Chem. Phys.* **78** 3297
[3] Perdew J P, Burke K and Ernzerhof M 1996 *Phys. Rev. Lett.* **77** 3865
[4] Kresse G and Furthmüller J 1996 *Phys. Rev. B* **54** 11169
[5] Kresse G and Joubert D 1999 *Phys. Rev. B* **59** 1758
[6] Wang H, Zhang L, Han J and E W 2018 *Comput. Phys. Commun.* **228** 178
[7] Zhang Y, Wang H, Chen W, Zeng J, Zhang L, Wang H and E W 2020 *Comput. Phys. Commun.* **252** 107206

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[8] Plimpton S 1995 *J. Comput. Phys.* **117** 1
[9] Henkelman G, Uberuaga B P, Jonsson H 2000 *J. Chem. Phys.* **113** 9901
[10] Nakano A 2008 *Comput. Phys. Commun.* **178** 280