Supporting Information

Electrically induced net-magnetization in $FePSe_3$ nanoribbons: The role of edge reconstructions

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The structure of $FePSe_3$ nanoribbons before being optimized is demonstrated in Fig. 1, where the arrangement of the edge Se atoms is in the armchair configuration and the edge Se atoms are unsaturated.

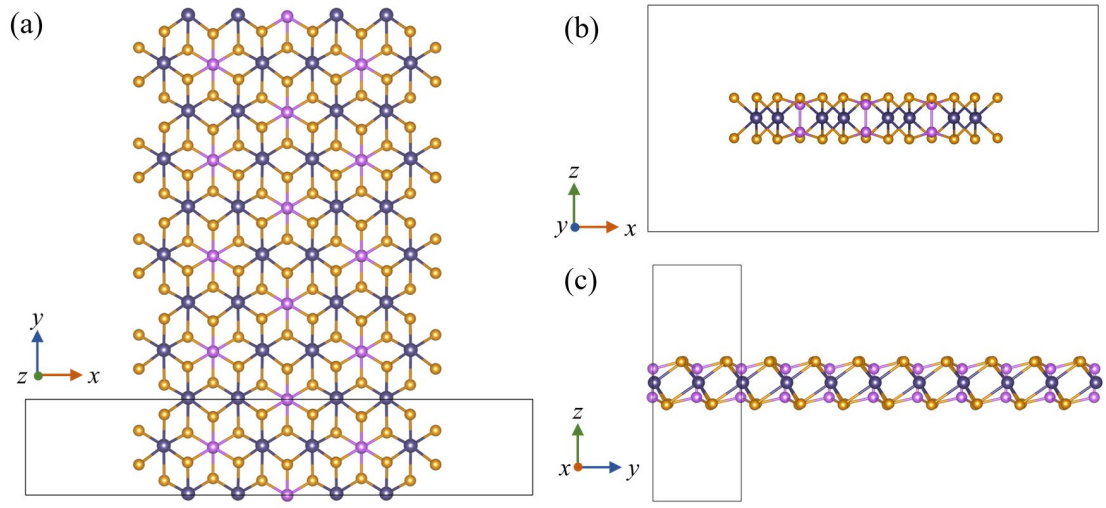

Figure 1. Top and side views of the unoptimized $FePSe_3$ nanoribbon structure. The dark blue, purple, and orange balls represent Fe atoms, P atoms and Se atoms, respectively.

The magnetic moments are mainly localized at the Fe atoms. The exchange interactions between the Fe atoms are mainly indirect via the intermediate Se atoms. The four magnetic configurations of the nanoribbon, namely ferromagnetic (FM), Néel-antiferromagnetic (AFM), zigzag-antiferromagnetic, and stripy-antiferromagnetic, are shown in Fig. 2. By comparing the total state energies of the magnetic configurations, it is shown that the zigzag AFM is the magnetic ground state. The calculated total energies relative to that of zigzag AFM are summarized in Table 1. The $FePSe_3$ nanoribbons energetically refers the zigzag-AFM magnetic configuration, and the energy difference between the ground state and FM is about 13.6 me V. The Calculations show that there is a local magnetic moment of 4 μ_B on each Fe atom. The magnetic anisotropy energy (MAE) is calculated as the energy difference according to the Force theorem [1], $E_{\mathrm{MAE}} = E_{[100]} - E_{[001]}$, where $E_{[100]}$ and $E_{[001]}$ are the total energies with the magnetizations along the transverse ([100]) and perpendicular ([001]) directions, respectively. It is shown that $FePSe_3$ nanoribbons have a perpendicular magnetic anisotropy, where the magnetization points out of the nanoribbon plane. Given the hybridization of Se-*p* and Fe-*d* orbitals, the Se atoms are spin polarized and with the direction opposite to that of their Fe neighbors, as shown in Fig3.

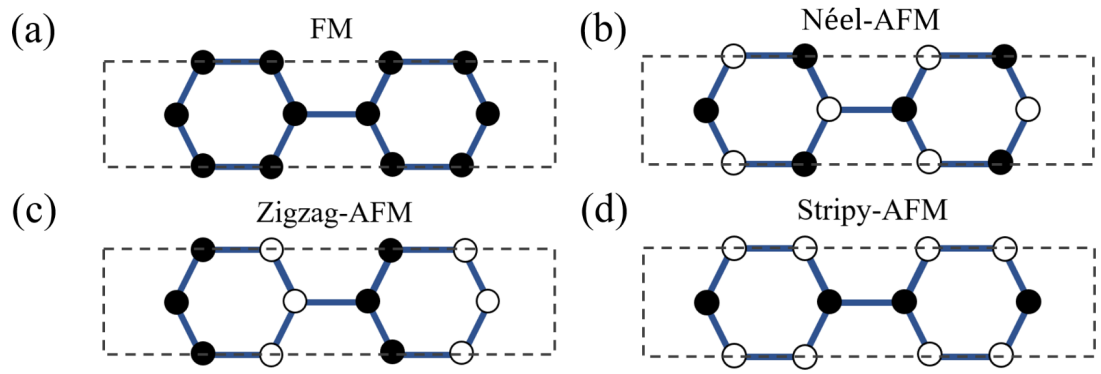

Figure 2. (a) - (d) Four different magnetic configurations of the $FePSe_3$ nanoribbon, where the black and white circles represent spin-up and spin-down, respectively.

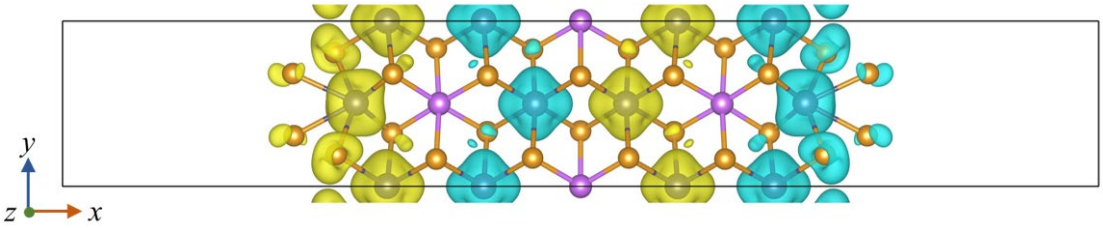

Figure 3. Spin density plots (3×10^{-3} eÅ^{-3}) of the $FePSe_3$ nanoribbon. The yello densities correspond to spin-up and blue to spin-down components.

Table 1. Total energy per atom (meV) of $FePSe_3$ nanoribbon with different magnetic configurations. The total energy of the Zigzag AFM magnetization is zero.

	FM	Zigzag-AFM	Néel-AFM	Stripy-AFM
ΔE(meV)	13.6	0	11.1	13.9

The evolution of the electronic structure of $FePSe_3$ nanoribbons to an external transverse electric field (E_x) of 0 ~ 0.6 V/Å is investigated. The direction of the electric field is in plane and perpendicular to the periodic direction. An intrinsic band gap of 0.83 eV is observed for both spin channels without an external electric field. The system is an antiferromagnetic semiconductor. As in Fig. 4, the band gap evolves when an external electric field is applied. When E_x = 0.1 V/Å, the spin-up bandgap increases to 1.07 eV, while the spin-down bandgap decreases to 0.63 eV. With further increase of the electric field, the spin-down bandgap rapidly vanishes. When E_x = 0.23 V/Å, similar to graphene nanoribbons, the $FePSe_3$ nanoribbons transform to a 100% spin-polarized half-metal, resulting in insulating behavior for spin-up and metallic behavior for spin-down. As the electric field continues increasing, the spin-up band gap also starts to decrease. As the electric field continues increasing, the conduction and valence bands cross the Fermi energy at E_x = 0.4 V/Å. When the E_x increases further, more electrons fill the spin-up states and fewer electrons fill the spin-down states, resulting in a linear increase of the total magnetic moment.

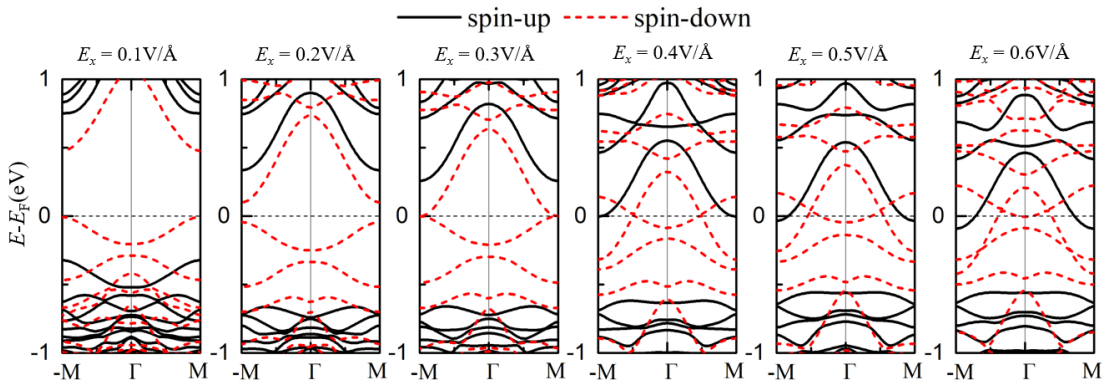

Figure 4. Energy bands under transverse electric fields of 0 ~ 0.6 V/ Å to the nanoribbon, where the black solid line is the spin-up state and the red dashed line is the spin-down state.

References

(1) Daalderop, G. H. O.; Kelly, P. J.; Schuurmans, M. F. H. First-Principles Calculation of the Magnetocrystalline Anisotropy Energy of Iron, Cobalt, and Nickel. *Phys. Rev. B* **1990**, *41* (17), 11919–11937.