Supporting Information for

"Electric field analyses on monolayer semiconductors: An

example of InSe"

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1. Effect of symmetrisation of the charge density

In the VASP code, a symmetry limitation (ISYM=2 for the PAW formalism) is used to achieve a more efficient, memory conserving symmetrisation of the charge density. However, such a default set-up will lead to serious problems when an electric field is applied. As shown in Fig. S1(a), an electric field induces the same charge polarization behavior on the upper and lower surfaces of InSe monolayer. Besides, the polarization is even independent of directions of fields. These results are contrary to the basic electrostatic law. In fact, we also find the same problem in other two-dimensional materials, such as MoS_2 monolayer and bilayer, see Fig. S2 and S3. Especially for the MoS_2 bilayer, due to the default set-up, an electric field will even induce an accumulation of charge density between two layers, which has been reported as a new result in pervious calculation.¹ Therefore, the symmetry set-up must be cancelled (for example, set ISYM=0) in the calculation when electric fields are applied. In this way, we can get reasonable change polarization behaviors and other properties induced by the electric fields.



Figure S1. Charge density difference in InSe monolayer with (a) and without (b) symmetry set-up under 1V/nm applied electric fields along +Z and -Z directions. Atoms color coding: dark brown for In and green for Se.



Figure S2. Charge density difference in MoS_2 monolayer (a) with and (b) without symmetry set-up under 1V/nm applied electric fields along +Z and -Z directions. Atoms color coding: black for Mo and pink for S.



Figure S3. Charge density difference in MoS_2 bilayer (a) with and (b) without the symmetry set-up under a 3V/nm applied electric field along +Z direction. Atoms color coding: black for Mo and pink for S.

2. Dielectric constant of InSe monolayer

As mentioned in the main text, the external field in the InSe monolayer reduces to about 1/8 from the vacuum. As pointed out in previous work,² the calculated dielectric constant for a supercell including 2D material dependents on the thickness of vacuum and can be written as ε^{ave} ,

$$\varepsilon^{ave} = \frac{d\varepsilon_z + (L_z - d)}{L_z} = 1 + \frac{1}{L_z} [d(\varepsilon_z - 1)]$$

Here, L_Z is the supercell size along Z direction. *d* is the thickness of a InSe monolayer (here, d=7.71Å). ε_Z is the true dielectric constant for InSe monolayer along Z direction. We can get the ε_Z from the slope of ε^{ave} vs. $1/L_Z$ as shown in Fig. S4, and accordingly $\varepsilon_Z \approx 9$. That is close to the reduction factor (8) in the InSe monolayer. Therefore, the reduction of the external field in InSe monolayer is consistent with the distribution of dielectric constant.



Figure S4. The calculated ε^{ave} versus $1/L_Z$ and their slope for InSe monolayer.

3. Position of the CBM and VBM under electric fields

Under electric fields, the position of CBM is insensitive to the electric field, see Fig. S5(b). While, the VBM moves towards the K point with increasing the electric field, see Fig. S5(a).



Figure S5. Band structure around the VBM (a) and CBM (b) under different effective electric fields.

4. Distribution of the CBM and VBM of the InSe monolayer



Figure S6. The distribution of the VBM and the CBM of the InSe monolayer which are reflected by a charge density isosurfaces of 5×10^{-3} a.u. (purple regions).

Reference

1. A. Ramasubramaniam, D. Naveh and E. Towe, *Phys. Rev. B: Condens. Matter*, 2011, **84**, 205325.

- 2. D. Wang, D. Han, X. B. Li, N. K. Chen, D. West, V. Meunier, S. B. Zhang and H.
- B. Sun, Phys. Rev. B: Condens. Matter, 2017, 96, 155424.