SUPPLEMENTARY INFORMATION

Dynamical amplification of electric polarization through nonlinear phononics in 2D SnTe

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Supplementary Note 1. Phonon dispersion in monolayer SnTe

In the ferroelectric ground-state configuration (one of the minima shown in Fig. 2(b)), our single phonon calculation at Gamma point reveals the $E_{u,x}$ and $E_{u,y}$ phonon frequencies at 1.33 THz and 1.39 THz, respectively. In our phonon dispersion as shown in Figs. 2a and Supplementary Figure 1, on the other hand, LO-TO correction gives rise to the stiffening of E_u (LO) phonon to 3.4 THz. Our results of the phonon frequencies without LO-TO splitting well correspond to the previous literature.[1] When we included the LO-TO splitting, one of the E_u mode shifts up, owing to the stiffening, but remaining bands produces similar dispersions as those reported in the literature.

Supplementary Note 2. Increased double well barrier by A_u phonon

The full range of the double well potential is presented in Supplementary Figure 2. Note that the barrier of the double well is substantially increased on the given atomic displacement toward A_u phonon mode.

Supplementary Note 3. Mode effective charge and Born effective charge

Here, we compare variation of mode effective charge and Born effective charge. In Supplementary Table 1 and 2, we explicitly summerized the diagonal (Z_x^x) and the off-diagonal parts (Z_x^z) of Born effective charge of each atom. Note that, at equilibrium (Q_{Au}=0), the diagonal part of Born effective charge of Sn and Te are almost exactly opposite. On the lattice distortions (along A_u modes or others), the Born effective charge of the Sn-Te pair can deviate from this exact oppositeness. It is notable that off-diagonal part of A_u mode effective charges at Q_{Au} = ±80mÅ give exactly opposite values, while off-diagonal part of Born effective charge shows asymmetrical changes as shown in Supplementary Table 2.

Supplementary Note 4. Nonlinear phonon interaction in group-IV monochalcogenide monolayer

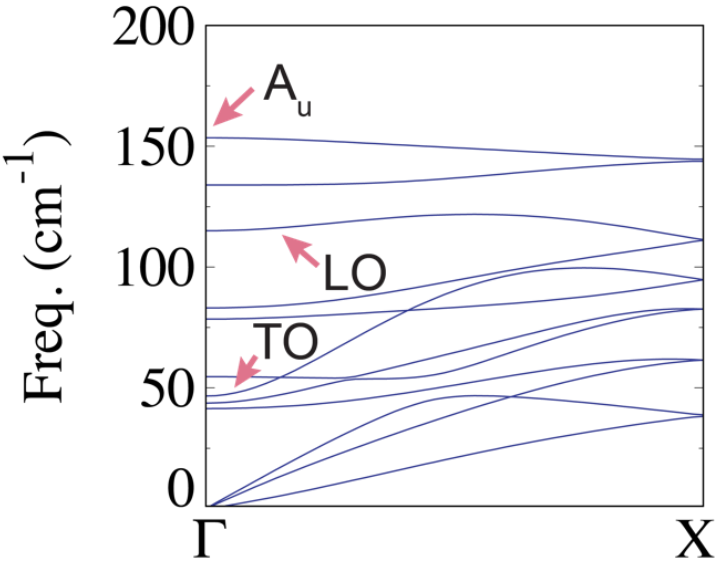
These characteristics features can be commonly observed from the family group-IV monochalcogenide monolayer, such as SnS and SnSe. These materials possesses similar electronic band structures and also very similar in-plane ferroelectricity, as depicted in Fig. 1 and Supplementary Figure 3.[2] To provide an explicit example, we repeated the potential energy surface of the monolayer SnS with lattice distortion along A_u and $E_{u,x}$ phonons, as summarized in Supplementary Figure 3. The potential energy surface for the monolayer SnS exhibits very similar fashion as the monolayer SnTe case, and thus the dynamical amplification of the electric polarization by nonlinear phonon interaction is also thought to be observed commonly from both SnS and SnTe.

$Z_x^x = dP_x/du_x$	Q_{Au} = -80mÅ	Q_{Au} = 0mÅ	Q_{Au} = 80mÅ
Sn1	6.54	6.23	5.91
Sn2	5.92	6.23	6.54
Te1	-6.34	-6.23	-6.11
Te2	-6.11	-6.23	6.34

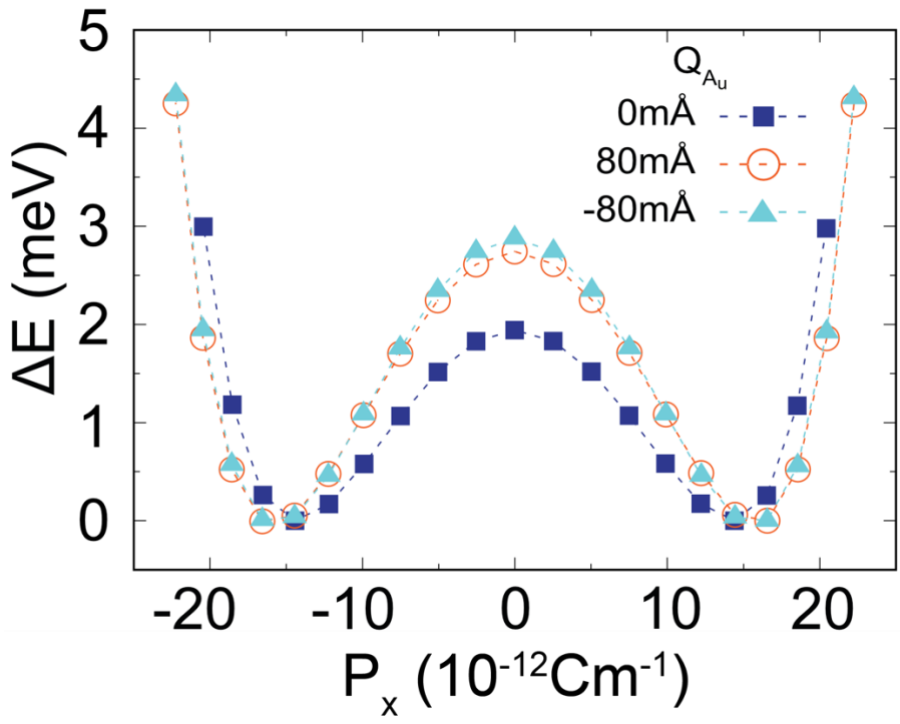
Supplementary Table 1. Diagonal part of the Born effective charge (Z^x_x) with respect to atomic displacement toward A_u mode.

$Z_x^z = dP_x/du_z$	Q_{Au} = -80mÅ	Q_{Au} = 0mÅ	Q_{Au} = 80mÅ
Sn1	0.024	0.042	0.057
Sn2	-0.057	-0.042	-0.024
Te1	0.016	0.000	-0.017
Te2	0.017	0.000	-0.016
Z^{Au}_x mode effective charge	-0.066	0.000	0.066

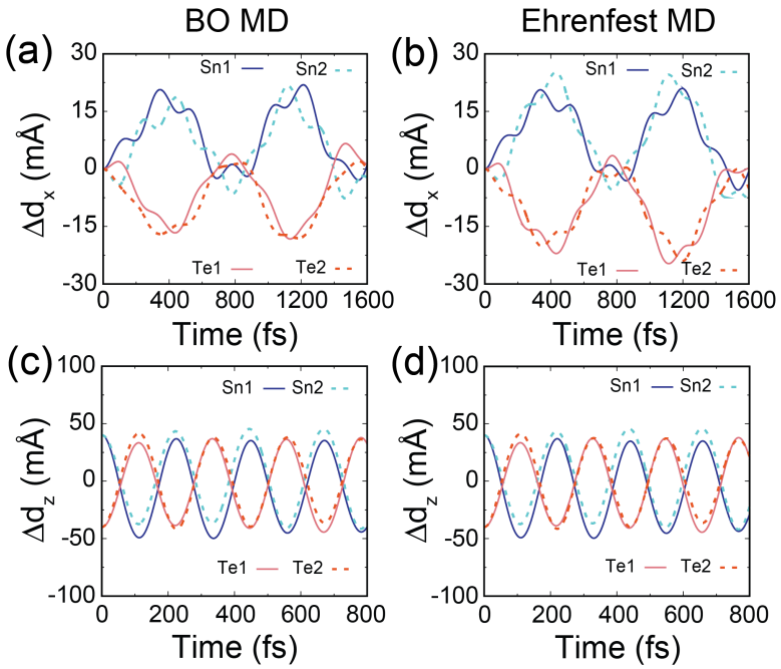
Supplementary Table 2. Off diagonal part of the Born effective charge (Z^z_x) and A_u mode effective charge (Z^{Au}_x) with respect to atomic displacement toward A_u mode.



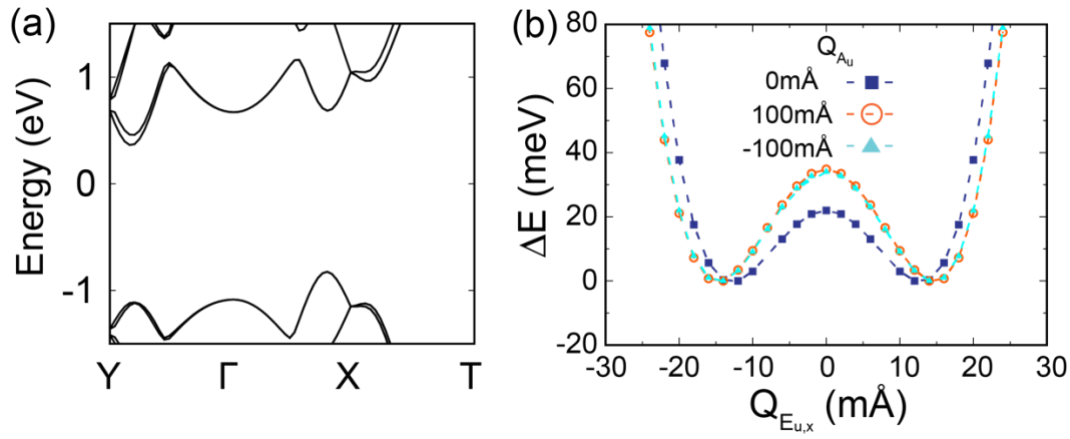
Supplementary Figure 1. Phonon dispersion of monolayer SnTe in cm^{-1} unit, including the LO-TO correction.



Supplementary Figure 2. The potential energy surface of the monolayer SnTe with given A_u mode (Q_{A_u}) distortions with respect to the in-plane electric polarization. Zoomed-in views near the potential well is depicted in Fig. 3d in the main text.



Supplementary Figure 3. Time-profile of atomic motions in the x-direction during (a) BO dynamics and (b) Ehrenfest dynamics initiated with Q_{Au}^{init} = 40mÅ. Time-profile of atomic motions in the z-direction during the (c) BO dynamics and (d) Ehrenfest dynamics initiated with Q_{Au}^{init} = 40mÅ. There is no significant difference description between Born-Oppenheimer and Ehrenfest molecular dynamics about nonlinear phonon interaction.



Supplementary Figure 4. (a) Band structure of ferroelectric monolayer SnS. (b) The potential energy surface of the monolayer SnS with respect to lattice displacements along the $E_{u,x}$ mode with given distortion along A_u mode (Q_{Au}).

Supplementary References

[1]Li, Y. et al. Promising thermoelectric properties and anisotropic electrical and thermal transport of monolayer SnTe. *Appl. Phys. Lett.* **114**, 083901 (2019).

[2]Xu, L., Yang, M., Wang, S. J. & Feng, Y. P. Electronic and optical properties of the monolayer group-IV monochalcogenides MX (M=Ge, Sn; X= S, Se, Te). *Phys. Rev. B* **95**, 235434 (2017).