Electronic Supplementary Information

Moiré-Pattern-Assisted Thermoelectric Enhancement in Tungsten Diselenide Bilayer

Naveen Kumar, Soumya Mondal, and Ayan Datta[†]

School of Chemical Sciences, Indian Association for the Cultivation of Science, Kolkata 700032, India

E-mail: spad@iacs.res.in

I. CONVERGENCE OF ELECTRONIC AND PHONON PROPERTIES

The convergence of the ground-state energy with respect to the k-point mesh and the convergence of the lattice thermal conductivity with respect to the q-point mesh are shown in **Fig. S1**. The ground-state energy is found to converge beyond a 7 × 7 × 1 k-point mesh, while the lattice thermal conductivity converges for q-point meshes denser than 15 × 15 × 1.

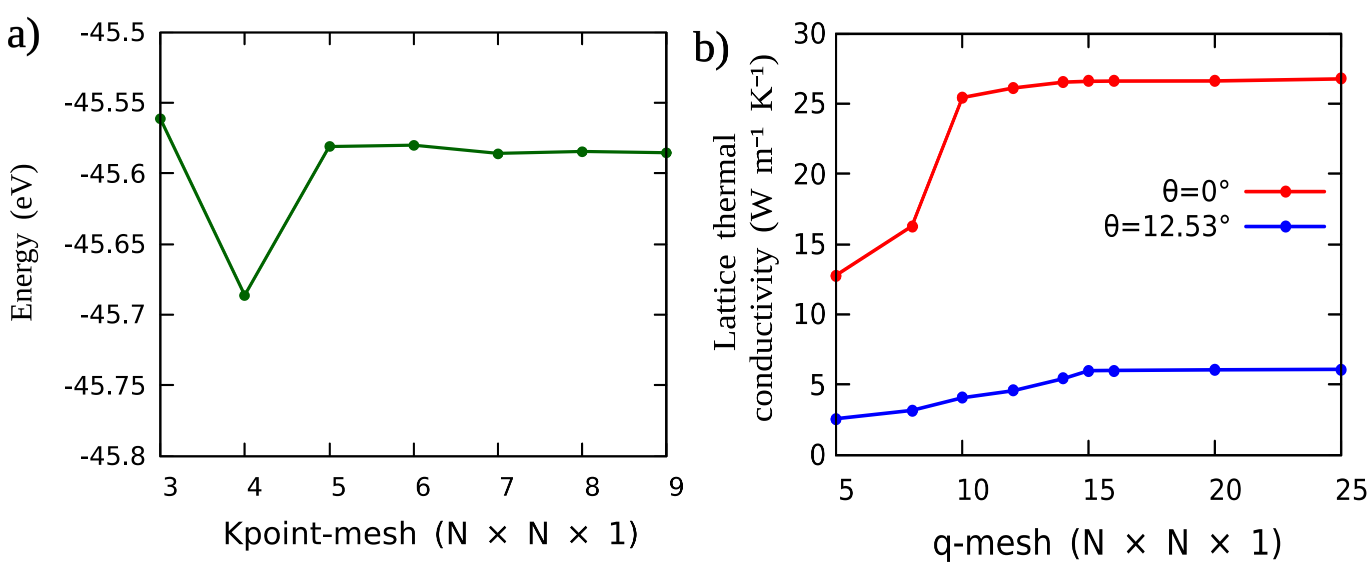

Figure S1. *Convergence of* ***(a)*** *electronic ground state energy with k-point mesh and* ***(b)*** *lattice thermal conductivity with q-point mesh.*

II. DEFORMATION POTENTIAL CALCULATIONS

The calculated deformation potential by applying uniaxial strain is shown in **Fig. S2**. The structures are isotropic, therefore, the deformation potential along both the in-plane directions is same.

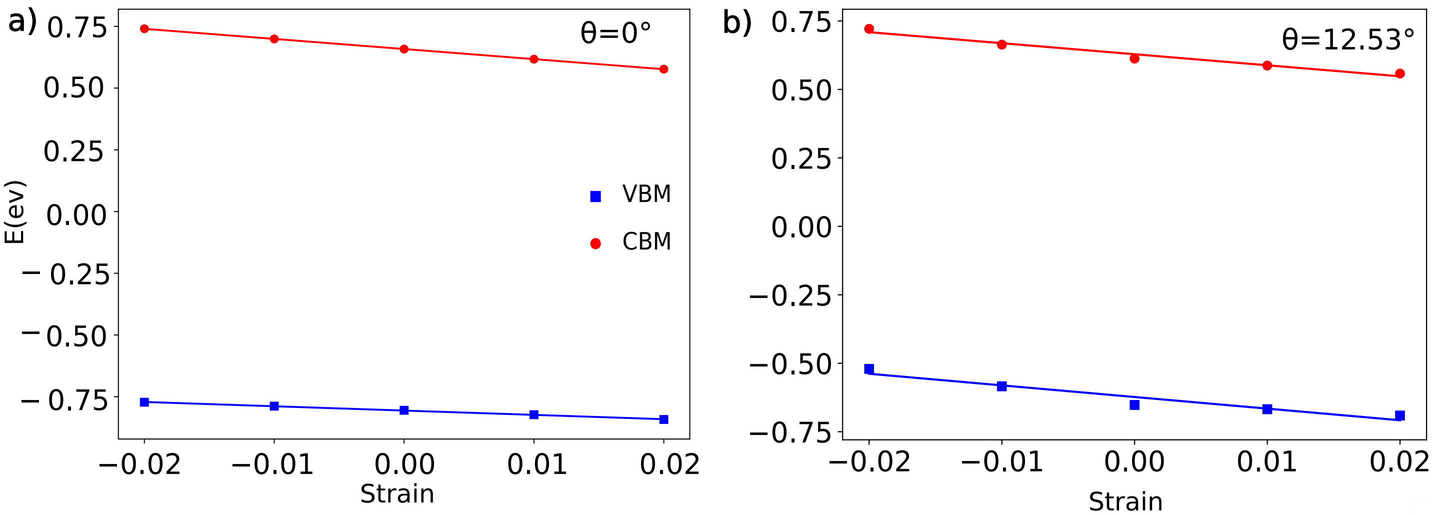

Figure S2. *Deformation potential calculations for* ***(a)*** *tungsten diselenide bilayer (0°)* ***(b)*** *12.53° twisted tungsten diselenide superlattice. VBM and CBM represent valence band maxima and conduction band minima, respectively.*

III. PHONON TRANSPORT PROPERTIES

The Grüneisen-parameter contour maps clearly demonstrate the strong influence of the moiré potential on the anharmonic behavior of twisted WSe_2 bilayer is shown in **S3** for all the three structures. The untwisted structure exhibits relatively uniform and moderate anharmonicity, while increasing twist leads to pronounced variations across the mini-Brillouin zone.

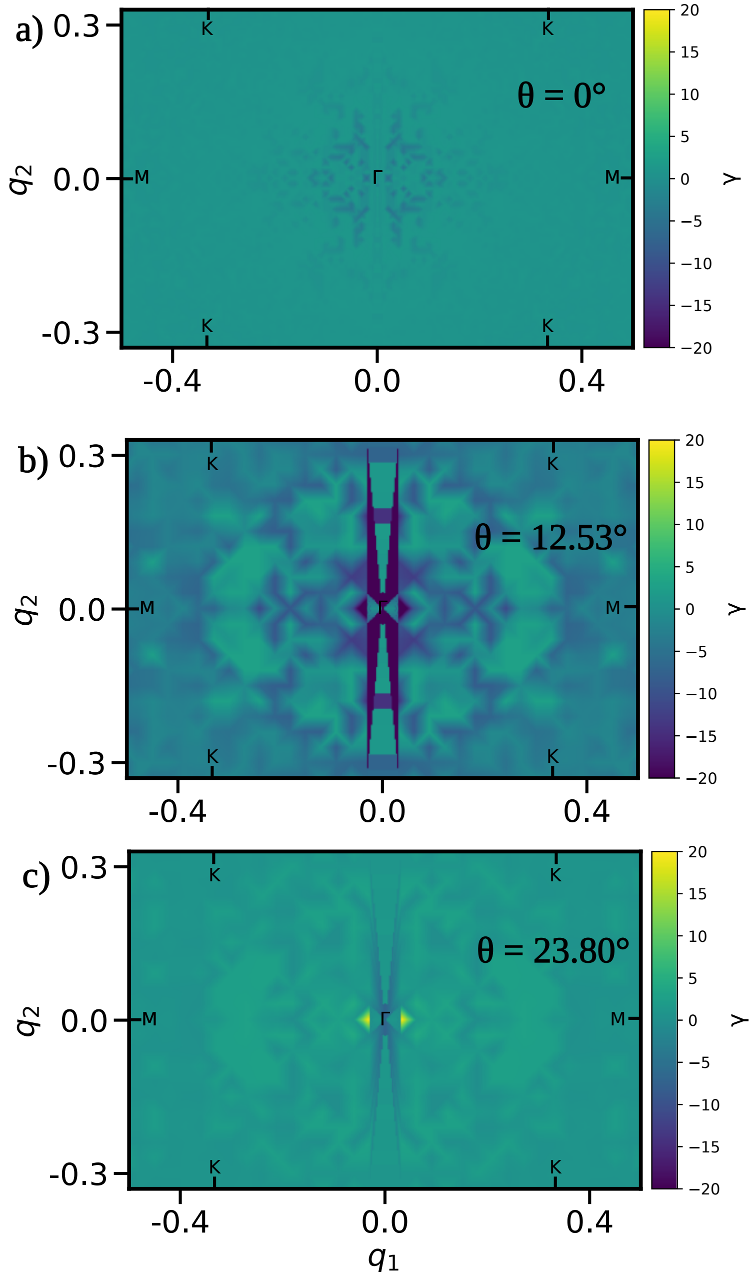

Figure S3. *Grüneisen-parameter contour maps plots for* ***(a)*** *tungsten diselenide bilayer (0°)* ***(b)*** *12.53° twisted, and* ***(c)*** *23.80° twisted tungsten diselenide superlattice. The range of color bar has been taken same for comparison.*

The variation of phonon lifetime with the low frequencies is shown in **S4**. It is observed that the untwisted bilayer configuration exhibits comparatively longer phonon relaxation times. In contrast, introducing a 12.53° twist markedly enhances phonon scattering, leading to a substantial reduction in relaxation time for phonons.

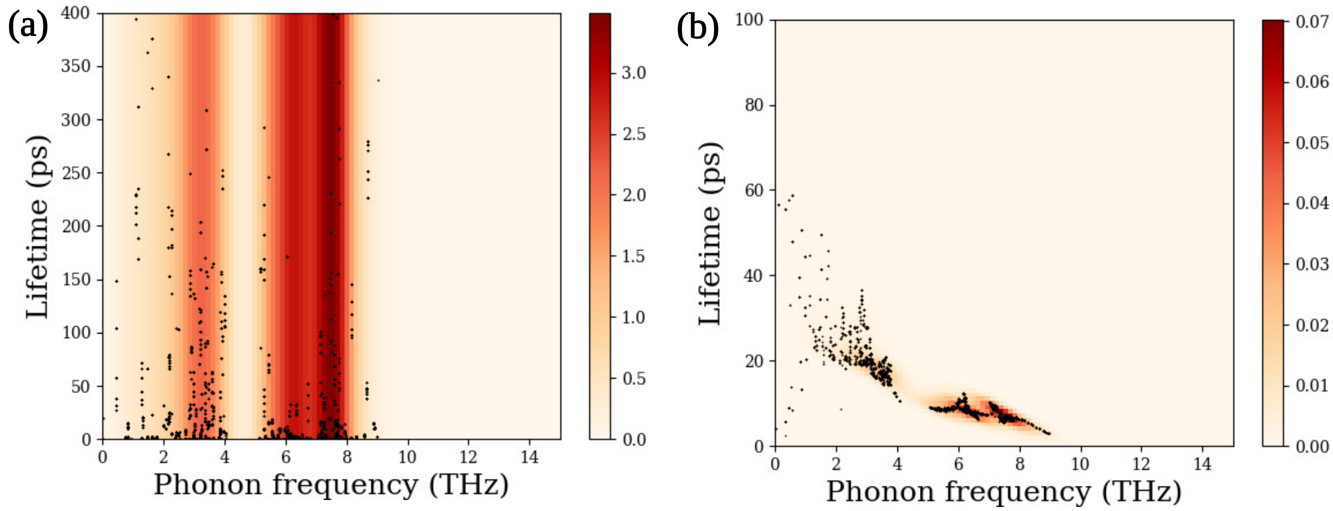

Figure S4. *Phonon lifetime vs frequency plots for* ***(a)*** *tungsten diselenide bilayer (0°)* ***(b)*** *12.53° twisted tungsten diselenide superlattice. The heatmap shows the probability density (how many phonon modes exist in that frequency–lifetime region) and the black-dots represent the actual data points for each phonon mode.*

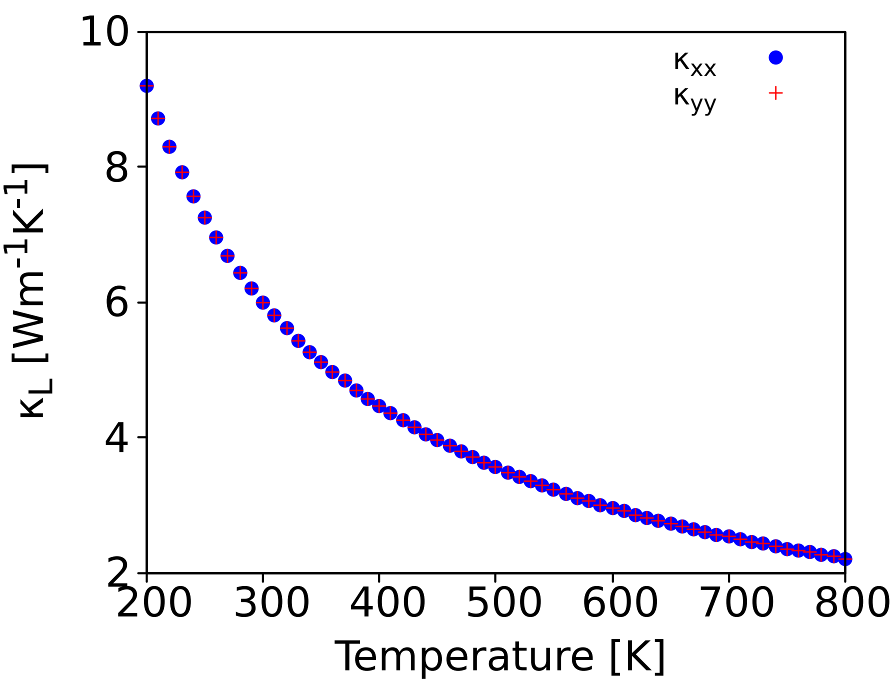

Figure S5. *Lattice thermal conductivity along the xx (solid blue circles) and yy (open red squares) directions as a function of temperature for 12.53° twisted tungsten diselenide superlattice. These two directions are effectively symmetry-equivalent with respect to in-plane thermal transport*

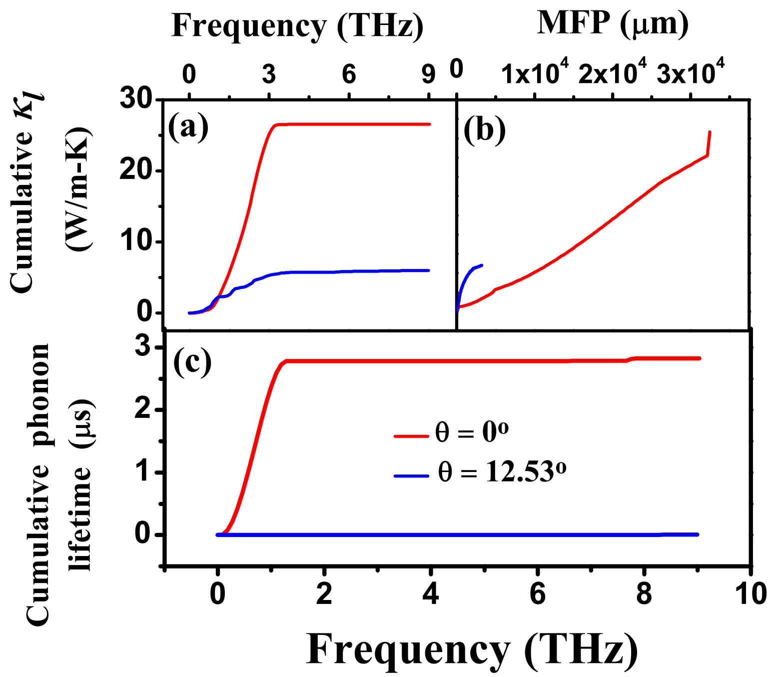

Figure S6. ***(a)*** *Cumulative lattice thermal conductivity vs frequency plot.* ***(b)*** *cumulative lattice thermal conductivity vs phonon mean-free path and* ***(c)*** *cumulative phonon lifetime vs frequency plot for tungsten diselenide bilayer (0°) and 12.53° twisted tungsten diselenide superlattice. The Y-axis for both figures (a) and (b) is kept identical.*

IV. SOC EFFECT ON ELECTRONIC BAND STRUCTURE

The comparison of electronic band structure for 0° twisted WSe_2 bilayer with and without spin orbit coupling effect is shown in **Fig. S7**. It clearly demonstrates that SOC induces a pronounced splitting of the electronic bands near the Fermi level.

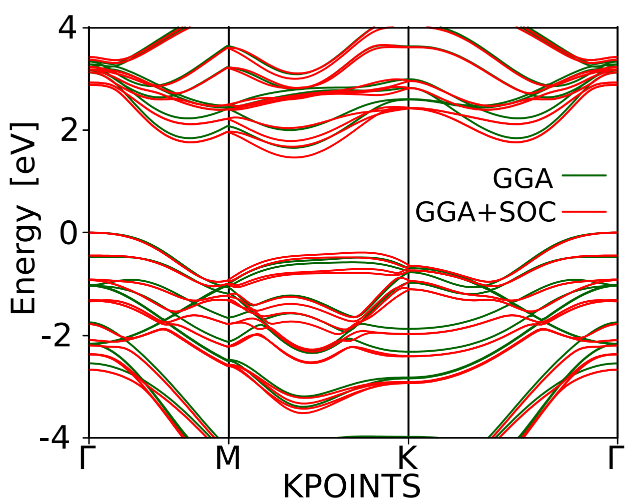

Figure S7. *Comparison of the electronic band structures of twisted WSe_2 bilayer calculated without spin–orbit coupling (GGA) and with spin–orbit coupling (GGA+SOC).*

V. ELECTRON-PHONON SCATTERING RELAXATION TIME

The temperature dependent electron-phonon scattering relaxation time for WSe_2 bilayer and 12.53° twisted WSe_2 bilayer for both types of charge carriers is shown in **Fig. S8**

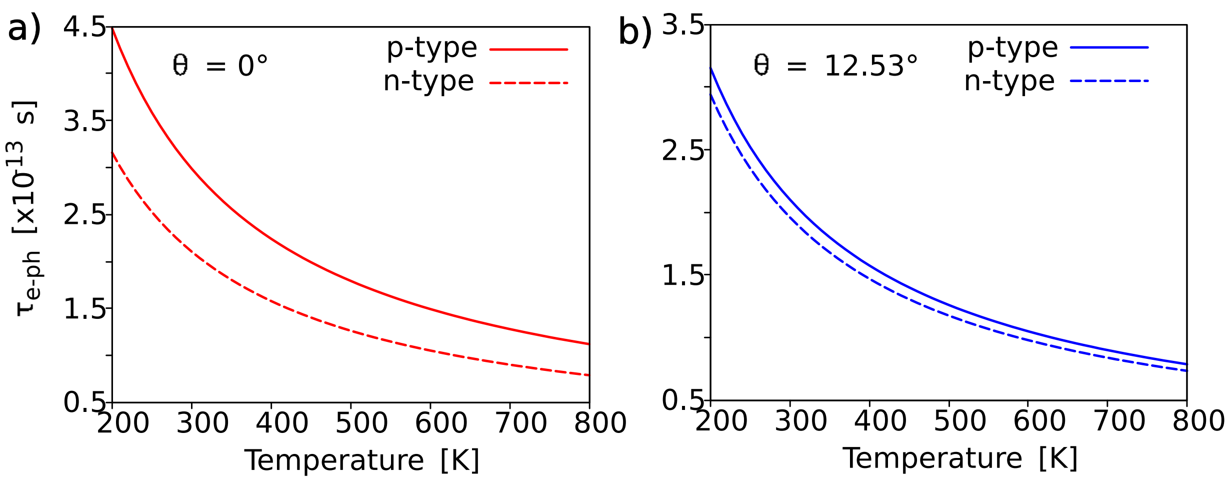

Figure S8. *Relaxation time for electron-phonon scattering of* ***(a)*** *tungsten diselenide bilayer (0°)* ***(b)*** *12.53° twisted tungsten diselenide superlattice. p-type and n-type refers to holes and electrons concentrations respectively.*