Electronic Supplementary Information for

Anisotropic Thermal Transport in Twisted Bilayer Graphene

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S1. Frequency dependent thermal conductivity analysis

The frequency dependent thermal conductivity in NEMD simulations is based on time Fourier transform of atomistic heat current, which is expressed as:

$$Q_{i\to j} = \frac{1}{2}\left\langle F_{ji}v_j - F_{ij}v_i \right\rangle$$

where F_{ij} is the force between atom i and j. v is the atom velocity. And the total heat current from group A to group B is defined as:

$$Q_{A\to B} = \sum_{i\in A}\sum_{j\in B}\left\langle F_{ji}v_j - F_{ij}v_i \right\rangle$$

Meanwhile, the time-correlation function is expressed as:

$$Q_{A\to B}(t) = \sum_{i\in A}\sum_{j\in B}\left\langle F_{ji}(0)v_j(t) - F_{ij}(0)v_i(t) \right\rangle$$

The time-correlation function related to Fourier transform by

$$Q_{A\to B}(\omega) = \int_{-\infty}^{+\infty} Q_{A\to B}(t)e^{i\omega t}dt$$

Thus, the spectrum thermal conductivity can be acquired based on the Fourier's law of thermal conductivity and the above spectrum heat current. Specific computational scheme can be seen in the work of Fan *et al*[1] and Zhou *et al*[2].

S2. Structure and parameters for twisted bilayer graphene

The angles of the commensurate rotation are based on the following equation if the periodicity of the crystal lattice is considered.

$$\cos\theta(m,n) = (3m^2 + 3mn + n^2/2)/(3m^2 + 3mn + n^2)$$

where m and n are the positive integral, respectively. In addition, the lattice constants and the atom numbers of those superlattice are also vary with the m and n. The number

of atoms is determined by the equation.

$$N = 4[(m+n)^2 + m(2m+n)]$$

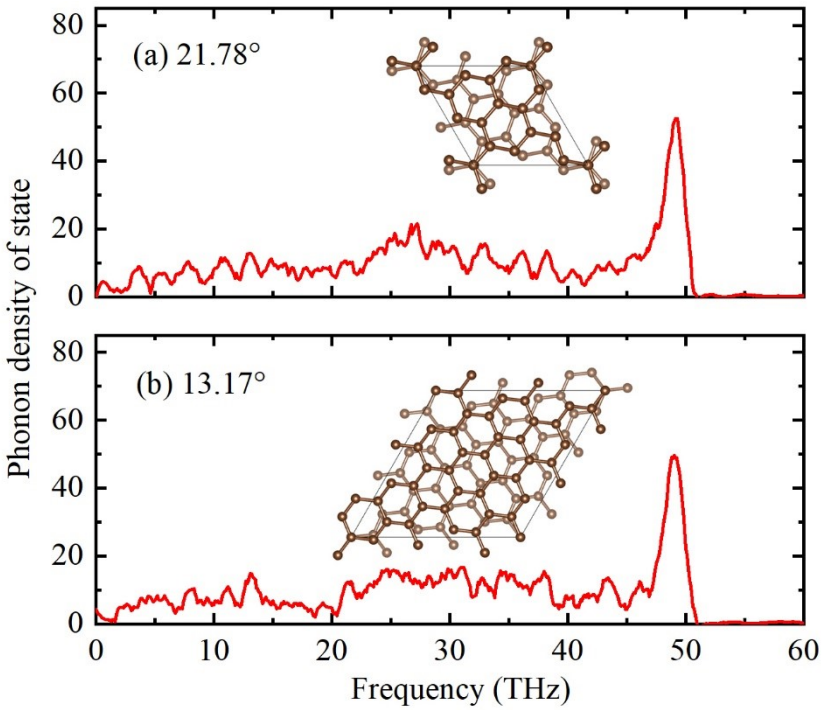
where N is number of atoms. And the lattice constants are expressed as:

$$L = \sqrt{3m^2n + 3mn + 1}a_0$$

where L is the lattice constant and a_0 is the initial lattice constant of bilayer graphene 2.46 Å. The corresponding values of the above parameters are shown in table S1. The geometric structures and phonon property with different twisting angles, including 21.78° and 13.17° are shown in Fig. S1.

m	*n*	Twisting angles	Lattice constant	Atom number
0	1	60	2.46	2
1	1	21.78	6.508	28
2	1	13.17	10.723	76
3	1	9.43	14.964	148

Table. S1 Parameters of twisted bilayer graphene



Figures. S1 Strusture and phonon density of state of twisted bilayer graphene with different twisting angles (a) 21.78° and (b) 13.17°.

Reference:

1. Z. Y. Fan, H. K. Dong, A. Harju and T. Ala-Nissila, *Phys Rev B*, 2019, **99**.

2. Y. G. Zhou and M. Hu, *Phys Rev B*, 2016, **93**.