Supporting Information of ”Moiré phonons in magic-angle twisted bilayer graphene”

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CONTENTS

I. Relaxed structure of twisted bilayer graphene (TBG) 2

II. Deep potential molecular dynamics 3

III. Low-frequency phonon modes and their symmetries 4

IV. Band structures under frozen phonon approximation 5

V. Lattice vectors and reciprocal lattice vectors of a commensurate supercell 7

VI. Flexural phonon modes of graphene 8

VII. Moiré phonon modes at different twisted angles 9

VIII. Electron-phonon coupling in some Γ point soft modes 10

References 16
I. RELAXED STRUCTURE OF TWISTED BILAYER GRAPHENE (TBG)

The atomic displacement vectors in the relaxed lattice structure of magic-angle TBG have been presented in Fig. S1, where the color coding and the vector fields indicate the amplitudes and directions of the atomic displacements. Inspection shows that after structural relaxation the interlayer distance in the AA region tends to be increased, leading to the out-of-plane corrugation. In particular, the interlayer distance at the AA point is as large as 3.62 Å, while at the AB/BA point it is 3.36 Å, consistent with previous report [1, 2]. This is clearly shown in Fig. S2, where the color coding denotes the in-plane variation of the interlayer distance in the moiré supercell. Moreover, the AA (AB/BA) region is contracted (expanded) after structural relaxation. The in-plane atomic displacement vectors wind around the AA point, forming a vortex-like pattern.

FIG. S2. The interlayer distance as a function of real-space coordinates (x, y) in the moiré superlattice.
II. DEEP POTENTIAL MOLECULAR DYNAMICS

FIG. S3. Validation of the neural network (NN) potential: (a) Comparison of total energies calculated from NN potential and DFT.deep-potential model prediction; (b-d) Comparison of forces calculated from NN potential and DFT in $x, y, z$ directions, respectively.

Primitive moiré supercells with different twist angles (from 21.79° to 4.41°) have been constructed to get complex enough local atomic environments [3]. The rotation center is chosen to be the center of the hexagon formed by the carbon atoms. With such a choice of rotation center, the $D_6$ symmetry of graphene is preserved for the TBG system. In order to describe the tiny lattice distortions of TBG during structural relaxations, we constructed several structures with small perturbative atomic displacements from ideal moiré pattern as the initial configurations for ab initio molecular dynamics (AIMD). For each configurations, we performed AIMD calculation for at least 100 fs to obtain plenty enough training data. The AIMD simulation have been carried out using the Vienna Ab initio Simulation Package (VASP) [4] with vDW-DF2 exchange-correlation functional [5]. $D_6$ symmetry is enforced in the AIMD simulation. Under periodic conditions, a vacuum layer of 20 Å in the $z$ direction has been set up. Then the reliable interatomic potential based on neural networks can be trained from DFT energies and forces, based on which we further studied the structural and phononic properties of TBG at smaller twist angles (including the magic angle $\theta \approx 1.08^\circ$) with
larger moiré primitive cells with the accuracy comparable to DFT calculations. Otherwise it would be extremely demanding to directly tackle with the structural properties of magic-angle TBG with traditional DFT method. The structural relaxation and force-constant matrix calculations were calculated by Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [6] with such NN potential[7]. Finally, based on the force-constant matrix, phonons were calculated by phonopy [8] package with a $2 \times 2$ moiré supercell. The accuracy of our NN potential has been validated by comparing the total energies and forces of various structures with DFT calculations, as shown in Fig. S3.

III. LOW-FREQUENCY PHONON MODES AND THEIR SYMMETRIES

![Image of phonon modes](image)

**FIG. S4.** The first 30 soft phonon modes at $\Gamma$

The real-space out-of-plane vibrational pattern of the first 30 soft optical phonon modes of magic-angle TBG at the high symmetry points $\Gamma$ and $M$ have been shown in Fig. S4 and Fig. S5 respectively. We further present the irreducible representations of the first 300
optical phonon modes at Γ, M, and K in S1.

**FIG. S5.** The first 30 soft phonon modes at M

**IV. BAND STRUCTURES UNDER FROZEN PHONON APPROXIMATION**

The energy bands with some typical soft phonon modes being frozen are shown in Fig. S6. In particular, in Fig. S6(a)-(c) we show the band structures with out-of-plane “octupolar” phonons assumed to be frozen, while in Fig. S6(d)-(f) we show the band structures with some “vortical” phonon modes being frozen (the phonon modes are shown in the insets). Our calculations indicate that the octupolar phonon modes tend to open up a gap at the charge neutrality point due to the broken $C_{2z}$ symmetry, while the “vortical” phonon modes tend to enhance intervalley scattering, which splits two Dirac points.
TABLE S1. The first 300 phonon modes in $\Gamma$, $M$, and $K$. Such representation is the point group in Schoenflies notation. The first 25 phonon modes have been shown in here, and the whole table could be seen in https://github.com/Simon-lxq/Phonon-symmetry

| eigen No. | $\Gamma$ | $M$ | $K$ |
|-----------|-----------|-----|-----|
| 1         | $D_6$     | $C_2$ | $C_6$ |
| 2         | $D_6$     | $C_2$ | $D_6$ |
| 3         | $D_6$     | $D_2$ | $D_6$ |
| 4         | $C_6$     | $C_2$ | $C_2$ |
| 5         | $C_2$     | $C_2$ | $D_6$ |
| 6         | $C_2$     | $C_2$ | $C_6$ |
| 7         | $D_2$     | $C_2$ | $D_6$ |
| 8         | $D_2$     | $D_2$ | $C_2$ |
| 9         | $D_3$     | $C_2$ | $C_6$ |
| 10        | $C_2$     | $C_2$ | $D_2$ |
| 11        | $C_2$     | $D_2$ | $C_2$ |
| 12        | $C_6$     | $C_2$ | $D_3$ |
| 13        | $D_2$     | $C_2$ | $C_6$ |
| 14        | $D_2$     | $C_2$ | $C_2$ |
| 15        | $D_3$     | $C_2$ | $C_2$ |
| 16        | $D_2$     | $C_2$ | $C_2$ |
| 17        | $D_2$     | $D_2$ | $C_1$ |
| 18        | $D_3$     | $C_2$ | $D_6$ |
| 19        | $C_2$     | $C_2$ | $C_6$ |
| 20        | $C_2$     | $C_2$ | $D_6$ |
| 21        | $C_6$     | $C_2$ | $D_6$ |
| 22        | $C_6$     | $D_2$ | $D_6$ |
| 23        | $C_2$     | $C_2$ | $D_3$ |
| 24        | $C_2$     | $C_2$ | $C_2$ |
| 25        | $D_2$     | $D_2$ | $D_3$ |
FIG. S6. The energy bands of typical phonon modes in high symmetry point Γ. (a)-(c) The band structure of the soft modes with the frequencies 0.0726 THz, 0.1634 THz, 0.2387 THz, respectively. The insets are the general patterns of out-of-plane vibrational amplitudes, where the average amplitudes is 0.05 Å.(d)-(f) The band structure of the soft modes with the frequencies 1.2184 THz, 2.1259 THz, 3.6348 THz, respectively. The insets are the general patterns of in-plane vibrational amplitudes.

V. LATTICE VECTORS AND RECIPROCAL LATTICE VECTORS OF A COMMENSURATE SUPERCELL

We consider studying a phonon mode at moiré wavevector \( \mathbf{q} \), which can be written as

\[
\mathbf{q} = \frac{q_1}{p_1} \mathbf{G}_1 + \frac{q_2}{p_2} \mathbf{G}_2
\]

(1)
where \((q_1, p_1)\) and \((q_2, p_2)\) are two pairs of co-prime integers. In order to plot the real-space vibrational pattern of the phonon mode at wavevector \(\mathbf{q}\), we need to construct a minimal real-space supercell with the superlattice vectors:

\[
\mathbf{R}_1 = n_{11}\mathbf{t}_1 + n_{12}\mathbf{t}_2 \\
\mathbf{R}_2 = n_{21}\mathbf{t}_1 + n_{22}\mathbf{t}_2
\]

(2)

We require \(\mathbf{R}_1 \cdot \mathbf{q} = 2\pi, \mathbf{R}_1 \cdot \mathbf{q} = 0\), such that

\[
\frac{n_{11}q_1}{p_1} + \frac{n_{12}q_2}{p_2} = 1 \\
\frac{n_{21}q_1}{p_1} + \frac{n_{22}q_2}{p_2} = 0
\]

(3)

And from Eq.(3) we obtain the expressions for the two reciprocal vectors of the commensurate supercell:

\[
\mathbf{G}_1^s = \frac{n_{22}\mathbf{G}_1 - n_{21}\mathbf{G}_2}{n_{11}n_{22} - n_{12}n_{21}} \\
\mathbf{G}_2^s = \frac{n_{11}\mathbf{G}_2 - n_{12}\mathbf{G}_1}{n_{11}n_{22} - n_{12}n_{21}}
\]

(4)

We find integer solutions of Eq.(3), and construct a supercell with minimal area. The area of the primitive supercell \(\Omega_S\) is expressed as:

\[
\Omega_S = (\mathbf{R}_1 \times \mathbf{R}_2) \cdot \hat{z} = (n_{11}n_{22} - n_{12}n_{21})\Omega_0
\]

(5)

VI. FLEXURAL PHONON MODES OF GRAPHENE

In this section, we show that the gapless flexural phonon modes in graphene are the origin of the low-frequency moiré modes in magic-angle twisted bilayer graphene. In Fig. S8 we show the flexural phonon modes of monolayer graphene at wavevectors \(q = n_1\mathbf{G}_1 + n_2\mathbf{G}_2\), where the vectors \(\mathbf{G}_1, \mathbf{G}_2, \mathbf{G}_1 + \mathbf{G}_2, \mathbf{G}_1 - \mathbf{G}_2, \mathbf{G}_1 + 2\mathbf{G}_2, 2\mathbf{G}_1 + \mathbf{G}_2\) is the basic vectors, \(\mathbf{G}_1, \mathbf{G}_2\) are the primitive reciprocal vectors of the moiré superlattice. If we consider the linear combination of these modes that mapped in two different monolayers, we may obtain the monopolar, dipolar, quadrupolar, and outupolar like phonon pattern, see Fig. S9. However, these linear superpositions of flexural modes are still significantly different from the calculated moiré phonon modes of magic-angle TBG (e.g., compare Fig. S9 with Fig. S4). This indicates that we need to consider the interlayer couplings, which would couple the
flexural modes from the two layers at different moire reciprocal vectors, such that the moire phonon patterns of TBG are significantly reconstructed compared to those of two decoupled graphene monolayers.

![Diagram of moire phonon modes](image)

FIG. S8. Long-wavelength flexural phonon modes of monolayer graphene at different wavevectors ($G_1, G_2, G_1 + G_2, G_1 - G_2, G_1 + 2G_2, 2G_1 + G_2$)

VII. MOIRÉ PHONON MODES AT DIFFERENT TWISTED ANGLES

In this section, we present the low-frequency moiré phonon modes at non-magic angles. As discussed above, those low-frequency moire phonon modes reported in our work actually originate from the linear superpositions of the flexural modes (“ZA modes”) of the two graphene monolayers. However, the interlayer couplings also play an important role, which would couple the flexural modes from the two layers at different moire reciprocal vectors, such that the moire phonon patterns of TBG are significantly reconstructed compared to those of two decoupled graphene monolayers. Therefore, from such a perspective, the magic angle is not so special, and such moire phonons can also emerge at non-magic angles. In particular, we have calculated the phonons of twisted bilayer graphene with the twist angle $\theta = 1.35, 1.78, 2.13, \text{and}3.15$ degrees, and the results are presented in Figs. S10, S11, S12, and S13, respectively. Clearly, those moire phonon modes at the magic-angle twisted bilayer
FIG. S9. Long-wavelength flexural phonon modes after linear combinations of the modes at different wavevectors in Fig. S8

graphene are also present at these non-magic angles.

What makes the magic-angle special is still the electronic flat bands, which would have strong charge fluctuations, large density of states, thus strong electron-phonon coupling effects. Therefore, these intriguing low-frequency moire phonon modes are more likely to become unstable around the magic angle due to the enhanced electron-phonon coupling effects.

VIII. ELECTRON-PHONON COUPLING IN SOME Γ POINT SOFT MODES

To further assess the effect of phonon modes on electron band structure, we choose several soft modes at Γ point and show their related results in this section. We start with
FIG. S10. Low-frequency moiré phonon modes for TBG with twisted angle $\theta = 1.35^\circ$

FIG. S11. Low-frequency moiré phonon modes for TBG with twisted angle $\theta = 1.78^\circ$
FIG. S12. Low-frequency moiré phonon modes for TBG with twisted angle $\theta = 2.13^\circ$

the standard electron-phonon coupling strengths\cite{9}:

$$\lambda_{nk} = 2N_F \sum_{m\nu\ell} \frac{|g_{mn\nu}(k, q)|^2}{\omega_{\nu\ell}} W_{m,k+q}$$

$$\lambda = \sum_{nk} \lambda_{nk} W_{nk}$$

(6)

where $m, n$ are electron band indices, $\nu$ is phonon band index, $q$ is the phonon wave vector, $\omega_{\nu\ell}$ is the frequency of the phonon mode, $N_F$ is the electron density of state per spin at Fermi level, and $W_{nk} = \delta(E - E_{nk})/N_F$ is the partial weight of the density of states. In Eq.(6), the electron-phonon matrix elements $g_{mn\nu}(k, q) = \langle m, k + q | \delta\hat{H}_{\nu\ell} | n, k \rangle$, where $\hat{H}_{\nu\ell}$ refers to the change in the electron Hamiltonian due to phonon mode $(q, \nu)$ under frozen phonon approximation, where the phonon mode vector is normalized to length of $l_{\nu\ell} = \sqrt{\hbar/(2M_C \omega_{\nu\ell})}$, with $M_C$ being the mass of a carbon atom.

Note that the definition in Eq.(6) counts the contribution of all phonon modes. To show the effect of a phonon mode $(q_0, \nu)$, some further treatment are applied to Eq.(6): the summation over $\nu$ in the first equation is removed, and the function in the summation in the first equation is replaced by $|g_{mn\nu}(k, q_0)|^2 W_{m,k+q}/\omega_{\nu\ell}$, i.e., we assume the phonon modes in the whole phonon bands to have the same effect as the effect of $(k, q_0)$. Also, we restrict the
indices $m$ and $n$ to be within the flat band subspace, because the flat bands are energetically well separated from the remote bands near the magic angle in relaxed structure. Then the electron phonon coupling strength defined in this way is

$$\lambda^{(q_0,\nu)}_{nk} = 2NF \sum_m |g_{mn}(k, q_0)|^2$$

$$\lambda^{(q_0,\nu)} = \sum_{nk} \lambda^{(q_0,\nu)}_{nk} W_{nk}$$

Note that the wave vector $q$ has already been integrated out in Eq.(7). We first plot $\sqrt{\sum_{mn} |g_{mn}(k, \nu)|^2}$ as a function of $k$ in Fig. S14. Here, the phonon mode chosen in Fig. S14 are all at $\Gamma$ point, so $q_0 = 0$. In the main text, the band structure in Fig. 4(b) suggests a relatively large electron-phonon coupling for the 18th optical phonon mode at $\Gamma$ with $\omega = 0.163$ THz, which opens up gaps at the Dirac points of the moire Brillouin zone. This is confirmed by the explicit calculations of the electron-phonon matrix elements in the flat-band subspace, i.e., the electron-phonon matrix element is largest at the $K$ and $K'$ points of the moire Brillouin zone ($\sim 0.17$ meV) for the 18th phonon mode.

Although the electron-phonon coupling matrix elements seem to be not so large compared to some of the crystalline materials without moire superlattice, their effects on the electronic flat bands in magic-angle TBG are still substantial due to the extremely narrow bandwidth.
FIG. S14. Heatmaps of $\sqrt{\sum_{mn}|g_{mn}(\mathbf{k},0)|^2}$ as a function of $\mathbf{k}$ for the 4th($0.043$ THz), 5th($0.046$ THz), 6th($0.046$ THz), 7th($0.063$ THz), 8th($0.063$ THz), 9th($0.073$THz), 12th($0.116$ THz), 15th($0.132$ THz), 18th($0.163$ THz), 21th($0.179$ THz), 22th($0.239$ THz), and 27th($0.253$ THz) Γ point phonon modes, in unit of eV. The dashed hexagon marks the first Brillouin zone, and the insets show the out-of-plane (normalized) vibrational amplitudes for the modes.
In Fig. S15 we present the Fermi energy dependence of $\lambda^{(0,\nu)}$ (defined in Eq. (7) for the 12 phonon modes shown in the insets of Fig. S14. We see that for the 4th mode and 18th mode, the maximal $\lambda^{(0,\nu)} \sim 0.2$ around the electronic van Hove singularities. Note that this is the contribution from just one particular phonon mode, without the summation over the mode index $\nu$, which indicates that the 4th and 18th phonon modes would have strong effects on the electronic band structures.

FIG. S15. Single-mode electron-phonon coupling strength $\lambda^{(q_0,\nu)}$ defined in Eq.(7) as a function of Fermi energy.
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