Graphene moiré superlattices are outstanding platforms to study correlated electron physics and superconductivity with exceptional tunability. However, robust superconductivity has been measured only in magic-angle twisted bilayer graphene (MA-TBG) and magic-angle twisted trilayer graphene (MA-TTG). The absence of a superconducting phase in certain moiré flat bands raises a question on the superconducting mechanism. In this work, we investigate electronic structure and electron-phonon coupling in graphene moiré superlattices based on atomistic calculations. We show that electron-phonon coupling strength $\lambda$ is dramatically different among graphene moiré flat bands. The total strength $\lambda$ is very large ($\lambda > 1$) for MA-TBG and MA-TTG, both of which display robust superconductivity in experiments. However, $\lambda$ is an order of magnitude smaller in twisted double bilayer graphene (TDBG) and twisted monolayer-bilayer graphene (TMBG) where superconductivity is reportedly rather weak or absent. We find that the Bernal-stacked layers in TDBG and TMBG induce sublattice polarization in the flat-band states, suppressing intersublattice electron-phonon matrix elements. We also obtain the nonadiabatic superconducting transition temperature $T_c$ that matches well with the experimental results. Our results clearly show a correlation between strong electron-phonon coupling and experimental observations of robust superconductivity.

Moiré materials have emerged as precisely tunable platforms to explore fascinating physical phenomena [1]. For example, twisted bilayer graphene (TBG) was predicted to have nearly flat bands at certain "magic angles" (MAs) [2,3], and experimentally shown to host correlated insulators and superconductivity [4,7]. Subsequently, various interaction-driven phases have been realized not only in magic-angle twisted bilayer graphene (MA-TBG) [8,13] but also in other graphene moiré superlattices [14-20]. In addition to electronic properties, extensive aspects of moiré physics have been actively explored, such as moiré excitons [21,22] and atomic-structure and phonon reconstructions [23,25].

While correlated insulating states are observed in many graphene moiré superlattices having flat bands, MA-TBG has been the only system to show robust superconductivity until recent experiments added magic-angle alternating-twist trilayer graphene (TTG) to the list [7,18-19]. In other systems, such as twisted double bilayer graphene (TDBG) and twisted monolayer-bilayer graphene (TMBG), superconducting phase appears to be rather weak or absent [15,16,26].

Naturally, the absence of robust superconductivity in some graphene moiré flat bands raises more questions on the superconducting mechanism. In particular, theoretical studies on MA-TBG have suggested that MA-TBG has strong electron-phonon coupling and phonon-mediated superconductivity is a strong candidate [27-30]. Also, strong electron-phonon coupling in MA-TBG is evidenced in experiments [25,31]. However, whether all the flat-band states in graphene moiré superlattices have such strong electron-phonon coupling is still unanswered, which has an important implication for the superconducting mechanism.

In this work, we investigate electronic structure and electron-phonon coupling of graphene moiré superlattices based on atomistic calculations. We show that electron-phonon coupling is strong ($\lambda > 1$) for TBG and TTG at their magic angles, but it is an order of magnitude weaker for TDBG and TMBG. We analyze such difference in $\lambda$ in terms of both density of states and matrix-element effects. We find that Bernal-stacked layers in TDBG and TMBG induce sublattice polarization in flat-band states, suppressing intersublattice electron-phonon matrix elements. Regardless of the total coupling strength, characteristic phonon modes are the same for all systems. We also calculate effects of vertical electric fields on electronic structure and electron-phonon coupling. Our calculations show that a correlation exists between strong electron-phonon coupling and experimental observations of robust superconductivity.

FIG. 1. Atomic relaxation patterns of (a) TBG with $\theta = 1.08^\circ$, (b) TTG with an alternating twist angle $\theta = 1.61^\circ$, (c) TDBG with $\theta = 1.35^\circ$, and (d) TMBG with $\theta = 1.25^\circ$. $\delta z$ is the deviation of atomic positions in the out-of-plane direction from the average value within each layer. Topmost two layers in (c) and bottom two layers in (c) and (d) are Bernal-stacked without twist.
We consider four graphene moiré superlattices at their respective magic angles, which is defined by the angle of the minimum bandwidth: TBG with $\theta = 1.08^\circ$, TTG with an alternating twist angle $\theta = 1.61^\circ$, TDBG with $\theta = 1.35^\circ$, and TMBG with $\theta = 1.25^\circ$. In (a), colored circles represent sublattice polarization (SP). TBG and TTG have exactly zero sublattice polarization. In (b), black lines show the total coupling strength. Red (blue) lines represent contributions of the inter- (intra-) sublattice electron-phonon coupling. In each case, the inset shows the average electron-phonon matrix element $V_{\text{ep}}$. (c), (d) Illustrations of how sublattice polarization suppresses intersublattice electron-phonon matrix elements. Blue arrows indicate electron-phonon matrix elements induced by phonon displacements denoted by green arrows. When sublattice polarization is nonzero in (d), the nearest-neighbor electron-phonon matrix elements are weakened.

We adopt atomistic approaches to calculate electrons and phonons in graphene moiré superlattices [28]. First, we calculate structural relaxations induced by variation of the stacking registry within moiré supercells, which have crucial effects on electronic structure of moiré flat bands [23] [32]. Equilibrium positions of all the carbon atoms are obtained by minimizing the sum of in-plane elastic energy and interlayer van der Waals binding energy [33] [34]. Then, electron states are obtained by diagonalizing atomistic tight-binding Hamiltonians with the Slater-Koster-type hopping integral parameterized for graphic systems [35] [36]. We calculate all the phonon modes in moiré supercells by diagonalizing dynamical matrices built from atomic force constants, which are the second derivatives of our total-energy function. With electron and phonon eigenstates, we compute electron-phonon matrix elements from changes in hopping amplitudes due to atomic displacements of phonon modes. From the above quantities, we can obtain electron-phonon coupling strength $\lambda$ and the Eliashberg function $\alpha^2 F(\omega)$ (See the Supplemental Material [37] for detailed descriptions of our methods).

Figure 1 shows our results of atomic relaxation patterns. $\delta z$ is the deviation of atomic positions in the out-of-plane direction from the average value of each layer. The average interlayer distances are about 3.40 Å between twisted layers and 3.35 Å between Bernal-stacked layers in TDBG and TMBG. In all systems, $\delta z$ is largest at AA stacking regions and has the opposite sign between twisted layers, except for TTG where the middle layer has zero $\delta z$ due to the symmetry and the other layers have large $\delta z$ in compensation. In our calculation, in-plane relaxations also occur in such a way to reduce the area of AA-stacked regions.

Figure 2(a) shows our calculated band structures and density of states (DOS) per spin. All the four systems have nearly flat bands and large DOS at their Fermi levels. Flat bands in TBG are the most archetypical in that nearly flat Dirac cones are located at the corners of the moiré Brillouin zone and isolated from the higher-energy bands. In TTG, the highly dispersing Dirac cone coexists with moiré flat bands. It comes from the outer graphene layers and is decoupled from the flat bands. In TDBG and TMBG, Dirac points at $K$ points are gapped because the absence of the inversion symmetry brings sublattice asymmetry. Our electronic structures are consistent with previous theoretical studies [28] [41] [44].

While TBG, TTG, TDBG, and TMBG at their respective magic angle have flat bands and large DOS at the Fermi level, we find a clear distinction of electron-phonon coupling strength $\lambda$ between the first two and last two systems. Figure 2(b) shows the total electron-phonon coupling strength of each system as a function of the Fermi energy. The most notable feature is that $\lambda$ is very strong in TBG and TTG with the maximum value reaching over 1, but it is an order of magnitude weaker in TDBG and TMBG. For TDBG and TMBG, $\lambda$ is less than 0.2 irrespective of the Fermi energy.

Such stark contrast in $\lambda$ originates partly from the differ-
FIG. 3. (a) Eliashberg function $\alpha^2 F(\omega)$, shown in red, at the half-filling energy of the hole-side in (upper left) TBG and (upper right) TTG, and the electron-side in (lower left) TDBG and (lower right) TMBG. Dashed blue lines denote frequency-integrated electron-phonon coupling strength $\lambda(\omega) = 2 \int_0^{\omega} \alpha^2 F(\omega')/\omega' d\omega'$. Insets show low-frequency ranges of $\alpha^2 F(\omega)$. (b) Phonon frequencies and momenta of the characteristic modes.

ence in the electronic density of states, but more crucially from the suppression of electron-phonon matrix elements in TDBG and TMBG. Insets in Fig. 2(b) show the average of electron-phonon matrix elements $V_{ep} = \lambda/N_F$, where $N_F$ is the electronic density of states at the Fermi energy $E_F$. While $V_{ep}$ in TBG and TTG is about 0.75 and 1.0 meV, respectively, it is below 0.3 meV in TDBG and TMBG. To explain why $V_{ep}$ is suppressed in TDBG and TMBG, we introduce sublattice polarization (SP) which measures the imbalance of sublattice weights of an electron state. For a given electron state $\psi_{nk}$, we define SP for each layer $l$ as

$$\text{SP}_l(\psi_{nk}) = \sum_{i \in A} |c_{nk,i}|^2 - \sum_{j \in B} |c_{nk,j}|^2, \quad (1)$$

where $c_{nk,j}$ is the tight-binding coefficient of $\psi_{nk}$ for an orbital centered at an atomic site $i$ and $A, B$ indicate two different sublattices. Then, the total SP, which is represented by colored circles in Fig. 2(a), is $\text{SP}(\psi_{nk}) = \sum_l |\text{SP}_l(\psi_{nk})|$. In TBG and TTG, the SP is exactly zero for all the electron states so they have the equal weights on two sublattices. In contrast, TDBG and TMBG, which both have Bernal-stacked layers, have nonzero SP and electrons have different sublattice weights within each layer, with signs of SP$_l$ alternating for different layers.

The presence of nonzero SP in the electronic structure of TDBG and TMBG critically weakens electron-phonon coupling strength. To illustrate this, we analyze the total electron-phonon coupling strength in terms of sublattice-dependent contributions. Figure 2(b) shows the total-electron-phonon coupling strength $\lambda$, and intersublattice ($\lambda^{AB} + \lambda^{BA}$) and intrasublattice ($\lambda^{AA} + \lambda^{BB}$) contributions as a function of the Fermi energy (see the Supplemental Material [37] for the formulas for inter- and intrasublattice $\lambda$). We find that, in all cases, the magnitude of the inter-sublattice contributions dominates the total coupling strength. This is because the strongest contribution comes from the electron-phonon matrix elements between the nearest neighbors, which belong to different sublattices [Fig. 2(c)]. In TBG and TTG, where SP is zero, electron wave functions have the same weights on both sublattices and the intersublattice matrix elements are strong. On the other hand, in TDBG and TMBG, nonzero SP suppresses the nearest-neighbor matrix elements [Fig. 2(c)], and the electron-phonon coupling becomes very weak.

Now, we analyze characteristic phonon modes that contribute to the total coupling strength. Figure 3(a) shows the Eliashberg functions $\alpha^2 F(\omega)$ and frequency-integrated electron-phonon coupling strength $\lambda(\omega) = 2 \int_0^{\omega} \alpha^2 F(\omega')/\omega' d\omega'$ of TBG and TTG (TDBG and TMBG) at the half-filling energy of the hole- (electron-) side. Regardless of the total coupling strength, characteristic phonon modes are the same for all systems. The largest portion of the total coupling strength comes from the in-plane optical modes near $\omega = 167 (197) \text{ meV}$ with phonon momentums at $q = K, K'$ (Γ). In addition, the interlayer breathing modes near $\omega = 10 \text{ meV}$, shown in the insets of Fig. 3(a), at $q = \Gamma$ also have sizable contribution due to their low energies. Table I summarizes mode-resolved electron-phonon coupling strength $\lambda_i$ for the three characteristic phonon modes shown in Fig. 3(b).

Since $E_F \approx 1-10 \text{ meV}$ and $\omega_{ph} \approx 10-200 \text{ meV}$, the adiabatic condition $\omega_{ph}/E_F < 1$ is extremely violated in graphene moiré superlattices. So the conventional McMillan formula does not apply. Instead, superconducting transition temperature $T_c$ in the nonadiabatic regime has nontrivial dependence on the electronic bandwidth[45][46]. If we ignore

| $\omega_i$ (meV) | TBG | TTG | TDBG | TMBG |
|-----------------|-----|-----|-------|-------|
| 10              | 0.297 | 0.233 | 0.064 | 0.037 |
| 167             | 0.914 | 0.743 | 0.026 | 0.045 |
| 197             | 0.648 | 0.532 | 0.018 | 0.030 |

| $D$ (meV) | 0.53 | 0.67 | 3.4 | 6.7 |
|----------|------|------|-----|-----|
| $T_c(\mu = 0.05)$ (K) | 3.45 | 3.76 | 10$^{-7}$ | 10$^{-6}$ |
| $T_c(\mu = 0.15)$ (K) | 3.33 | 3.55 | 0.0 | 0.0 |
the dispersion of phonon modes and the energy dependence of the electron DOS, an explicit $T_c$ formula for the half-filled bands can be derived as \[ T_c = \prod_i \left( \frac{\omega_i D}{\omega_i + D} \right)^{\lambda_i / \lambda} \exp \left( -\frac{1 + \lambda_i}{\lambda_i - \mu^*} \right), \] (2)

where $D$ is the half bandwidth, $\omega_i$ and $\lambda_i$ are the energy and electron-phonon coupling strength of the $i$th phonon mode, $\lambda = 2 \sum_i \lambda_i D / (\omega_i + D)$ is the mass renormalization constant, and $\mu^* = \mu / (1 + \mu \sum_i \ln(1 + D / \omega_i) \lambda_i / \lambda)$ is the Coulomb pseudopotential. We calculate $T_c$ at the half-filling Fermi energy of the hole- (electron-) side flat bands for TBG and TTG (TDBG and TMBG). Our results for $T_c$ are summarized in Table I and show good agreement with experimental observations in TBG and TTG \([7, 18, 19]\).

Lastly, we investigate electric-field effects on electronic structure and electron-phonon coupling. We consider a vertical electric field in our tight-binding Hamiltonian by adding an electrostatic energy term $\Delta H = eEz$, where $e > 0$ is the elementary charge, $z$ is the $z$ coordinate of an atom, and $Ez$ represents the total electric field consisting of external and induced electric fields.

Figures 4(a) and 4(b) show electronic structures and electron-phonon coupling strengths under the vertical electric field, respectively. In our calculation, TBG is nearly insensitive to the vertical electric field, except that two Dirac points at $K$ are split in energy because of the potential energy difference between two layers. Consequently, electron-phonon coupling strength is hardly affected by the electric field. In TTG, the electric field primarily affects highly dispersive monolayer Dirac bands so that they are pushed away from the flat bands. This makes the flat-band states more dispersive, reducing the density of states and, accordingly, decreasing the electron-phonon coupling strength. However, $V_{\text{ep}}$ is not affected by electric fields.

On the other hand, the electronic structures in TDBG and TMBG are much more sensitive to electric fields. In both systems, flat bands are split into the electron and hole sides, and the electron-side flat bands become narrower. Nevertheless, TDBG under electric fields shows very weak $\lambda$ because the sublattice polarization still suppresses $V_{\text{ep}}$. In more detail, we note a slight increase of $\lambda$ in the electron-side flat bands of TMBG as the electric field polarizes the electron-side states to the top monolayer where sublattice polarization is weaker.

In TBG and TTG, superconductivity often appears near spin-valley flavor-polarized correlated phases at certain integer fillings \([12, 13, 18, 19, 47]\). Several studies have suggested that such correlated phases compete with the superconductivity \([48, 49]\). In particular, experiments with controlled metallic gates have shown that superconductivity survives even after correlated phases disappear as a result of the enhanced screening from metallic gates \([50]\). Flavor polarization, if any, may weaken phonon-mediated superconductivity in two ways. First, flavor polarization may raise or lower band energies depending on their flavors, increasing the band width and reducing the density of states, which reduces the overall electron-phonon coupling strength. Second, flavor polarization may lift energy degeneracy of electron states with opposite momenta and opposite spins, for instance, $|k, \uparrow\rangle$ and $| -k, \downarrow\rangle$, which disturbs Cooper-pair formation for spin-singlet superconductivity. Effects of flavor polarization and electron correlation need to be studied for a full phase diagram of twisted graphene layers. In addition, further studies should include doping-dependent band dispersions and a more accurate description of Coulomb matrix elements \([51, 55]\).

To summarize, we have studied electronic structure and electron-phonon coupling in graphene moiré superlattices based on atomistic approaches. We find that total electron-phonon coupling is strong for MA-TBG and MA-TTG, but is an order of magnitude weaker for TDBG and TMBG, where the Bernal-stacked layers induce sublattice polariza-
tion, suppressing the nearest-neighbor electron-phonon matrix elements. Our results provide a deeper understanding into the electron-phonon coupling in graphene moiré superlattices, showing a correlation between strong electron-phonon coupling and experimental observations of robust superconductivity.

This work was supported by NRF of Korea (Grants No. 2020R1A2C3013673 and No. 2017R1A5A1014862) and KISTI supercomputing center (Project No. KSC-2020-CRE-0335). Y.W.C. acknowledges support from NRF of Korea (Global Ph.D. Fellowship Program NRF-2017H1A2A1042152).

---

* hj.choi@yonsei.ac.kr

[1] E. Y. Andrei, D. K. Efetov, P. Jarillo-Herrero, A. H. MacDonald, K. F. Mak, T. Senthil, E. Tutuc, A. Yazdani, and A. F. Young, The marvels of moiré materials, Nat. Rev. Mater. 6, 201 (2021).

[2] R. Bistritzer and A. H. MacDonald, Moiré bands in twisted double-layer graphene, Proc. Natl. Acad. Sci. U.S.A. 108, 12233 (2011).

[3] J. M. B. Lopes dos Santos, N. M. R. Peres, and A. H. Castro Neto, Graphene Bilayer with a Twist: Electronic Structure, Phys. Rev. Lett. 99, 256802 (2007).

[4] S. Shaller, S. Sharma, E. Kandelaki, and O. A. Pankratov, Electronic structure of turbostratic graphene, Phys. Rev. B 81, 165105 (2010).

[5] E. Suárez Morell, J. D. Correa, P. Vargas, M. Pacheco, and Z. Baticic, Flat bands in slightly twisted bilayer graphene: Tight-binding calculations, Phys. Rev. B 82, 121407(R) (2010).

[6] Y. Cao, V. Fatemi, A. Demir, S. Fang, S. L. Tomarken, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras, R. C. Ashoori, and P. Jarillo-Herrero, Correlated insulator behaviour at half-filling in magic-angle graphene superlattices, Nature (London) 556, 80 (2018).

[7] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, Unconventional superconductivity in magic-angle graphene superlattices, Nature (London) 556, 43 (2018).

[8] Y. Choi, H. Kim, Y. Peng, A. Thomson, C. Lewandowski, R. Polski, Y. Zhang, H. S. Arora, K. Watanabe, T. Taniguchi, J. Alica, and S. Nadj-Perge, Correlation-driven topological phases in magic-angle twisted bilayer graphene, Nature (London) 589, 536 (2021).

[9] K. P. Nuckolls, M. Oh, D. Wong, B. Lian, K. Watanabe, T. Taniguchi, B. A. Bernevig, and A. Yazdani, Strongly correlated Chern insulators in magic-angle twisted bilayer graphene, Nature (London) 588, 610 (2020).

[10] H. Polshyn, J. Zhu, M. A. Kumar, Y. Zhang, F. Yang, C. L. Tscharth, M. Serlin, K. Watanabe, T. Taniguchi, A. H. MacDonald, and A. F. Young, Electrical switching of magnetic order in an orbital Chern insulator, Nature (London) 588, 66 (2020).

[11] M. Serlin, C. L. Tscharth, H. Polshyn, Y. Zhang, J. Zhu, K. Watanabe, T. Taniguchi, L. Balents, and A. F. Young, Intrinsicit quantized anomalous Hall effect in a moiré heterostructure, Science 367, 900 (2020).

[12] D. Wong, K. P. Nuckolls, M. Oh, B. Lian, Y. Xie, S. Jeon, K. Watanabe, T. Taniguchi, B. A. Bernevig, and A. Yazdani, Cascade of electronic transitions in magic-angle twisted bilayer graphene, Nature (London) 582, 198 (2020).

[13] U. Zondiner, A. Rozen, D. Rodan-Legrain, Y. Cao, R. Queiroz, T. Taniguchi, K. Watanabe, Y. Oreg, F. von Oppen, A. Stern, E. Berg, P. Jarillo-Herrero, and S. Ilani, Cascade of phase transitions and Dirac revivals in magic-angle graphene, Nature (London) 582, 203 (2020).

[14] G. Chen, A. L. Sharpe, P. Gallagher, I. T. Rosen, E. J. Fox, L. Jiang, B. Lyu, H. Li, K. Watanabe, T. Taniguchi, J. Jung, Z. Shi, D. Goldhaber-Gordon, Y. Zhang, and F. Wang, Signatures of tunable superconductivity in a trilayer graphene moiré superlattice, Nature (London) 572, 215 (2019).

[15] X. Liu, Z. Hao, E. Khalaf, J. Y. Lee, Y. Ronen, H. Yoo, D. H. Najafabadi, K. Watanabe, T. Taniguchi, A. Vishwanath, and P. Kim, Tunable spin-polarized correlated states in twisted double bilayer graphene, Nature (London) 583, 221 (2020).

[16] Y. Cao, D. Rodan-Legrain, O. Rubies-Bigorda, J. M. Park, K. Watanabe, T. Taniguchi, and P. Jarillo-Herrero, Tunable correlated states and spin-polarized phases in twisted bilayer–bilayer graphene, Nature (London) 583, 215 (2020).

[17] G. Chen, A. L. Sharpe, E. J. Fox, Y.-H. Zhang, S. Wang, L. Jiang, B. Lyu, H. Li, K. Watanabe, T. Taniguchi, Z. Shi, T. Senthil, D. Goldhaber-Gordon, Y. Zhang, and F. Wang, Tunable correlated Chern insulator and ferromagnetism in a moiré superlattice, Nature (London) 579, 56 (2020).

[18] Z. Hao, A. M. Zimmermann, P. Ledwith, E. Khalaf, D. H. Najafabadi, K. Watanabe, T. Taniguchi, A. Vishwanath, and P. Kim, Electric field–tunable superconductivity in alternating-twist magic-angle trilayer graphene, Science 371, 1133 (2021).

[19] J. M. Park, Y. Cao, K. Watanabe, T. Taniguchi, and P. Jarillo-Herrero, Tunable strongly coupled superconductivity in magic-angle twisted trilayer graphene, Nature (London) 590, 249 (2021).

[20] S. Chen, M. He, Y.-H. Zhang, V. Hsieh, Z. Fei, K. Watanabe, T. Taniguchi, D. H. Cobden, X. Xu, C. R. Dean, and M. Yankowitz, Electrically tunable correlated and topological states in twisted monolayer–bilayer graphene, Nat. Phys. 17, 374 (2021).

[21] E. M. Alexeiev, D. A. Ruiz-Tijerina, M. Danovich, M. J. Hamer, D. J. Terry, P. K. Nayak, S. Ahn, S. Pak, J. Lee, J. I. Sohn, M. R. Molas, M. Koperski, K. Watanabe, T. Taniguchi, K. S. Novoselov, R. V. Gorbachev, H. S. Shin, V. I. Fal’ko, and A. I. Tartakovskii, Resonantly hybridized excitons in moiré superlattices in van der Waals heterostructures, Nature (London) 567, 81 (2019).

[22] K. Tran, G. Moody, F. Wu, X. Lu, J. Choi, K. Kim, A. Rai, D. A. Sanchez, J. Quan, A. Singh, J. Embley, A. Zepeda, M. Campbell, T. Autry, T. Taniguchi, K. Watanabe, N. Lu, S. K. Banerjee, K. L. Silverman, S. Kim, E. Tutuc, L. Yang, A. H. MacDonald, and X. Li, Evidence for moiré excitons in van der Waals heterostructures, Nature (London) 567, 71 (2019).

[23] H. Yoo, R. Engelke, S. Carr, S. Fang, K. Zhang, P. Cazeaux, S. H. Sung, R. Hovden, A. W. Tseng, T. Taniguchi, K. Watanabe, G.-C. Yi, M. Kim, M. Luskin, E. B. Tadmor, E. Kaxiras, and P. Kim, Atomic and electronic reconstruction at the van der Waals interface in twisted bilayer graphene, Nat. Mater. 18, 448 (2019).

[24] J. Quan, L. Linhart, M.-L. Lin, D. Lee, J. Zhu, C.-Y. Wang, W.-T. Hsu, J. Choi, J. Embley, C. Young, T. Taniguchi, K. Watanabe, C.-K. Shih, K. Lai, A. H. MacDonald, P.-H. Tan, F. Lishib, and X. Li, Phonon renormalization in reconstructed MoS2 moiré superlattices, Nat. Mater. 20, 1100 (2021).

[25] A. C. Gadelha, D. A. A. Ohlberg, C. Rabelo, E. G. S. Neto, T. L. Vasconcelos, J. L. Campos, J. S. Lemos, V. Ornelas, D. Mi-
[26] S. Chen, M. He, Y.-H. Zhang, V. Hsieh, Z. Fei, K. Watanabe, T. Taniguchi, D. H. Cobden, X. Xu, C. R. Dean, and M. Yankowitz, Electrically tunable correlated and topological states in twisted monolayer–bilayer graphene, Nat. Phys. 17, 374 (2021).

[27] F. Wu, A. H. MacDonald, and I. Martin, Theory of Phonon-Mediated Superconductivity in Twisted Bilayer Graphene, Phys. Rev. Lett. 121, 257001 (2018).

[28] Y. W. Choi and H. J. Choi, Strong electron-phonon coupling, electron-hole asymmetry, and nonadiabaticity in magic-angle twisted bilayer graphene, Phys. Rev. B 98, 241412(R) (2018).

[29] T. J. Peltonen, R. Ojajärvi, and T. T. Heikkilä, Mean-field theory for superconductivity in twisted bilayer graphene, Phys. Rev. B 98, 220504(R) (2018).

[30] B. Lian, Z. Wang, and B. A. Bernevig, Twisted Bilayer Graphene: A Phonon-Driven Superconductor, Phys. Rev. Lett. 122, 257002 (2019).

[31] H. Polshyn, M. Yankowitz, S. Chen, Y. Zhang, K. Watanabe, T. Taniguchi, C. R. Dean, and A. F. Young, Large linear-temperature resistivity in twisted bilayer graphene, Nat. Phys. 15, 1011 (2019).

[32] N. N. T. Nam and M. Koshino, Lattice relaxation and energy band modulation in twisted bilayer graphene, Phys. Rev. B 96, 075311 (2017).

[33] T. Nakamichi and T. Ando, Conductance of crossed carbon nanotubes, J. Phys. Soc. Jpn. 70, 1647 (2001).

[34] P. Moon and M. Koshino, Energy spectrum and quantum Hall effect in twisted bilayer graphene, Phys. Rev. B 85, 195458 (2012).

[35] See Supplemental Material for (i) detailed descriptions of our atomistic calculation methods for relaxed atomic structures, phonon spectra, electronic structures, and electron-phonon matrix elements, and (ii) formulas for sublattice analysis of electron-phonon interaction, which includes Refs. [38–40].

[36] A. A. Maradudin and S. H. Vosko, Symmetry properties of the normal vibrations of a crystal, Rev. Mod. Phys. 40, 1 (1968).

[37] A. Marek, V. Blum, R. Johanni, V. Hlav, B. Lang, T. Auckenthaler, A. Heinze, H.-J. Bungartz, and H. Lederer, The ELPA Library - Scalable Parallel Eigenvalue Solutions for Electronic Structure Theory and Computational Science, J. Condens. Matter Phys. 26, 213201 (2014).
Supplemental Material:  
Dichotomy of Electron-Phonon Coupling in Graphene Moiré Flat Bands  
Young Woo Choi and Hyoun Joon Choi*  
Department of Physics, Yonsei University, Seoul 03722, Korea  

In this supplemental material, we present detailed descriptions of our atomistic calculation methods for relaxed atomic structures, phonon spectra, electronic structures, and electron-phonon matrix elements, which are also used in our previous works on electron-phonon coupling in twisted bilayer graphene [S1] and electronic structure in twisted double bilayer graphene [S2]. In addition, we also present formulas for sublattice analysis of electron-phonon interaction.

S1. Structural Relaxation of Twisted Graphene Layers

The twist angle $\theta$ for a commensurate moiré supercell of graphene layers is given by

$$\cos \theta = \frac{M^2 + N^2 + 4MN}{2(M^2 + N^2 + NM)} \quad (S1)$$

for a pair of integers $(M, N)$ defining the supercell. In the case of twisted bilayer graphene (TBG), for instance, we consider supercell lattice vectors

$$t^{(1)}_1 = Na_1 + Ma_2,$$

$$t^{(1)}_2 = -Ma_1 + (M + N)a_2 \quad (S2)$$

of the bottom layer and

$$t^{(2)}_1 = Ma_1 + Na_2,$$

$$t^{(2)}_2 = -Na_1 + (M + N)a_2 \quad (S3)$$

of the top layer before twist. Here $a_1 = a(\sqrt{3}/2, -1/2)$ and $a_2 = a(\sqrt{3}/2, +1/2)$ are the primitive lattice vectors of graphene, and $a = 2.46 \text{ Å}$ is the lattice parameter. Then, a commensurate supercell is formed by rotating the bottom and top layers by $-\theta/2$ and $\theta/2$, respectively, so that the lattice vectors of the supercell are

$$t_1 = R(-\theta/2)t^{(1)}_1 = R(\theta/2)t^{(1)}_2,$$

$$t_2 = R(-\theta/2)t^{(2)}_1 = R(\theta/2)t^{(2)}_2 \quad (S4)$$

Here $R(\theta)$ is the clockwise rotation by $\theta$ around the hollow center of the graphene hexagon.

Other twisted graphene layers are generated in a similar way. Alternating-twist trilayer graphene (TTG) is formed by adding a graphene layer on the top of TBG, where the third layer is aligned with the first layer. For twisted double bilayer graphene (TDBG), two Bernal-stacked bilayers are twisted instead of two monolayers. Twisted mono-bilayer graphene (TMBG), then, is formed by removing the topmost layer from TDBG, resulting in a monolayer twisted on the top of a Bernal-stacked bilayer.

After generating rigidly twisted graphene layers, we relax atomic positions by minimizing the total energy as a function of all atomic positions. The total energy $U$ per a moiré supercell consists of the in-plane elastic energy and interlayer van der Waals binding energy:

$$U = \frac{1}{2} \sum_{i} \sum_{\alpha, \beta} C_{\text{MLG}}^{\alpha \beta} \Delta \tau^{i\alpha}_0 \Delta \tau^{i\beta}_0 + \frac{1}{2} \sum_{l \neq l'} \sum_{i, j} V_{\text{KC}}(\tau^{i\alpha}_{0l} - \tau^{i\alpha}_{0l'}). \quad (S5)$$

Here $\tau^{i\alpha}_{0l} = R_p + \tilde{R}$ is the position of the $i$th atom in the $l$th layer in the $p$th moiré supercell located at $R_p$, $\alpha$ is the cartesian index ($\alpha = x, y, z$), $\Delta \tau^{i\alpha}_0 = \tau^{i\alpha}_p - \tilde{\tau}^{i\alpha}_p$ is the deviation from the nonrelaxed position $\tilde{\tau}^{i\gamma}_p$, and $C_{\text{MLG}}^{\alpha \beta}$ are force constants between two atoms in the same layer up to the fourth-nearest neighbors. The explicit form of the force constants is given by Eq. (S5) of Ref. S3 and their values are shown in the column ‘4NNFC diagonal fit to GGA’ of Table 3 of Ref. S3, obtained by fitting to ab initio phonon dispersion calculations of monolayer graphene (MLG). The interlayer binding energy is calculated using Kolmogorov-Crespi (KC) potential $V_{\text{KC}}$ that depends on interlayer atomic registry [S4]. The explicit form of the KC potential is given by Eq. (3) of Ref. S4, where values of parameters are $C_0 = 15.71$ meV, $C_2 = 12.29$ meV, $C_4 = 4.933$ meV, $C = 3.030$ meV, $\delta = 0.578$ Å, $\lambda = 3.629$ Å$^{-1}$, $A = 10.258$ meV, and $z_0 = 3.34$ Å, as given in Ref. S4. We use a conjugate gradient method to minimize the total energy $U$ as a function of all atomic coordinates within the moiré supercell, whose total degrees of freedom are about 30 000 in the case of TBG with $\theta \sim 1^\circ$.

S2. Atomistic Calculation of Phonons in Twisted Graphene Layers

We calculate atomic force constants by taking the second derivatives of Eq. (S5) with respect to atomic positions:

$$C_{\text{pia}, \text{p}'; \beta}^{\alpha \beta} = \partial^2 U / \partial \tau^{\alpha}_{\text{pia}} \partial \tau^{\beta}_{\text{p'} \beta}. \quad (S6)$$

As the in-plane elastic energy is quadratic, the in-plane force constants are just those of monolayer graphene,

$$C_{\text{pia}, \text{p}'; \beta}^{\alpha \beta} = C_{\text{MLG}}^{\alpha \beta}. \quad (S7)$$
which are not changed by lattice relaxation. The interlayer force constants are the second derivatives of the KC potential,
\[ C_{p_{i\sigma},p'_{j\beta}} = -\frac{\partial^2 V_{KC}}{\partial x_{\alpha} \partial x_{\beta}} (\tau_{p_{i\sigma}} - \tau_{p'_{j\beta}}), \tag{S8} \]
which are evaluated at relaxed atomic positions.

From the force constants, the dynamical matrix is
\[ D_{i\alpha,j\beta}(q) = \sum_p e^{iq \cdot R_p} C_{0i\alpha,pj\beta}/M_C \tag{S9} \]
for a phonon wave vector \( q \), where \( M_C \) is the mass of a carbon atom. Then, we solve the phonon eigenvalue problem
\[ \omega^2 q_{\nu,i} = \sum_{j,\beta} D_{i\alpha,j\beta}(q) q_{\nu,j\beta} \tag{S10} \]
for the irreducible Brillouin zone to get the phonon energy \( \omega_{q\nu} \) and polarization vector \( e_{q\nu,i} \) of the \( \nu \)th phonon mode. The phonons in the rest of the Brillouin zone are obtained from the symmetry relations [S5]. We use the ELPA library to diagonalize the dynamical matrix efficiently [S6].

### S3. Atomistic Tight-Binding Method for Electronic Structure

We use a single-orbital tight-binding approach to calculate electronic structures of twisted graphene layers. Hamiltonian matrix elements are
\[ \hat{H} = \sum_{p_{i\sigma},p'_{j\sigma}} t(\tau_{p_{i\sigma}} - \tau_{p'_{j\sigma}}) |\phi_{i\sigma}; R_p \rangle \langle \phi_{j\sigma}; R_p|, \tag{S11} \]
where \( |\phi_{i\sigma}; R_p \rangle \) is a carbon \( p_{\sigma} \)-like orbital centered at \( \tau_{p_{i\sigma}} \). Here we drop the layer index on \( \tau_{p_{i\sigma}} \) so the index \( i \) sweeps all atoms in all layers in the \( p \)th moiré supercell at \( R_p \). We use the Slater-Koster-type hopping integral
\[ t(d) = V_{pp\sigma}^p e^{-(d_{\sigma\sigma}/\delta)} \left\{ 1 - (d_z/d)^2 \right\} + V_{pp\sigma}^0 e^{-(d_{\alpha\beta}/\delta)} (d_z/d)^2, \tag{S12} \]
where \( d \) is the displacement vector between two orbitals. The hopping energy \( V_{pp\sigma}^0 = -2.7 \text{ eV} \) is between in-plane nearest neighbors separated by \( a_0 = a/\sqrt{3} = 1.42 \text{ Å} \), and \( V_{pp\sigma}^0 = 0.48 \text{ eV} \) is between two vertically aligned atoms at the distance \( d_0 = 3.35 \text{ Å} \). Here \( \delta = 0.184a_0 \) is chosen to set the magnitude of the next-nearest-neighbor hopping amplitude to be 0.1V$_{pp\sigma}^0$ [S7,S8]. We use the cutoff distance \( d_c = 10 \text{ Å} \), beyond which the hopping integral is negligible.

### S4. Electron-Phonon Matrix Elements

The total electron-phonon coupling strength \( \lambda \) is
\[ \lambda_{nk} = 2N_F \sum_{m}\frac{|g_{mn\nu}(k,q)|^2}{\omega_{q\nu}} W_{m+n+k}, \tag{S13a} \]
\[ \lambda = \sum_{nk} \lambda_{nk} W_{nk}, \tag{S13b} \]
where \( N_F \) is the electron density of states per spin at the Fermi energy \( E_F \), \( W_{nk} = \delta(E_F - \varepsilon_{nk})/N_F \) is the partial weight of the density of states contributed by the \( n \)th electronic state \( |nk \rangle \) with wave vector \( k \) and energy \( \varepsilon_{nk} \), and \( g_{mn\nu}(k,q) = \langle m+k+q|\delta_n \hat{H} |nk \rangle \) is the electron-phonon matrix element between \( |nk \rangle \) and \( |m+n+k \rangle \) mediated by the \( q \)th phonon mode of wave vector \( q \) and frequency \( \omega_{q\nu} \). We calculate \( W_{nk} \) using the linear tetrahedron method [S9]. In our tight-binding approach, we expand electron and phonon eigenstates in localized basis sets to obtain the electron-phonon matrix element [S1]
\[ g_{m+n\nu}(k,q) = \sum_{i,\alpha,p} e_{q\nu,i}^\dagger \frac{\partial t(\tau_{oi} - \tau_{pj})}{\partial x_{\alpha}} |e_{q\nu,i} \rangle \langle e_{q\nu,j}| \times \{ e^{ik \cdot R_p} c_{m+n+k,q,i}^\dagger c_{nk,j}^\dagger + e^{-i(q+k) \cdot R_p} c_{m+n+k,q,j}^\dagger c_{nk,i} \}, \tag{S14} \]
where \( q_{\nu} = \sqrt{\hbar/(2M_C \omega_{q\nu})} \) is the length scale of phonon mode \( (q_{\nu}) \) and \( c_{nk,i} \) is the tight-binding coefficient of the electron state \( |nk \rangle \). The Eliashberg function can also be calculated from the matrix elements as
\[ \alpha^2 F(\omega) = N_F \sum_{m+n+k} |g_{mn\nu}(k,q)|^2 \times W_{nk} W_{m+n+k} \delta(\omega - \omega_{q\nu}), \tag{S15} \]
which shows the energy dependence of the momentum-averaged electron-phonon coupling.

### S5. Formulas for Sublattice Analysis of Electron-Phonon Coupling

In Eq. (S14), each term in the summation depends on two atomic sites indexed by \( i \) and \( j \), so the matrix element can be decomposed into intra- and inter-sublattice contributions depending on whether \( i \) and \( j \) sites belong to the same or different sublattices,
\[ g_{mn\nu}(k,q) = g_{mn\nu}^{AA}(k,q) + g_{mn\nu}^{BB}(k,q) \]
\[ + g_{mn\nu}^{AB}(k,q) + g_{mn\nu}^{BA}(k,q), \tag{S16} \]
where
\[ g_{mnS_iS_2}(k,q) = \sum_{i \in S_i, j \in S_2} \sum_{\alpha,p} e_{q\nu,i}^\dagger \frac{\partial t(\tau_{oi} - \tau_{pj})}{\partial x_{\alpha}} \times \{ e^{ik \cdot R_p} c_{m+n+k,q,i}^\dagger c_{nk,j}^\dagger + e^{-i(q+k) \cdot R_p} c_{m+n+k,q,j}^\dagger c_{nk,i} \}, \tag{S17} \]
is the matrix element between sublattices \( S_1 \) and \( S_2 \).

In terms of the decomposed matrix elements, we can also identify sublattice-dependent contributions to the total coupling strength,
\[ \lambda = \lambda^{AA} + \lambda^{BA} + \lambda^{AB} + \lambda^{mixed}, \tag{S18} \]
where

$$\lambda^{S_1, S_2} = 2N_F \sum_{nmkq} \left| \frac{g_{mn}^{S_1, S_2}(k, q)}{\omega_{qn}} \right|^2 W_{nk} W_{mk+q}$$  \hspace{1cm} (S19)

is the coupling strength between sublattices $S_1$ and $S_2$, and the mixed term contains the rest of the matrix elements $\{g_{mn}^{S_1, S_2}(k, q)\}^* g_{mn}^{S_1', S_2'}(k, q)$ with $(S_1, S_2) \neq (S_1', S_2')$. As shown in Fig. 2(b) of the main text, the inter-sublattice terms $\lambda^{AB/BA}$ have the dominant contributions to the total coupling strength. The intra-sublattice terms $\lambda^{AA/BB}$ are an order of magnitude weaker than $\lambda^{AB/BA}$. We note that $\lambda^{mixed}$ is not always positive-valued, but its magnitude is only comparable to $\lambda^{AA/BB}$ so it also has negligible contribution to the total coupling strength.

* h.j.choi@yonsei.ac.kr

[S1] Y. W. Choi and H. J. Choi, Strong electron-phonon coupling, electron-hole asymmetry, and nonadiabaticity in magic-angle twisted bilayer graphene, Phys. Rev. B 98, 241412(R) (2018).

[S2] Y. W. Choi and H. J. Choi, Intrinsic band gap and electrically tunable flat bands in twisted double bilayer graphene, Phys. Rev. B 100, 201402(R) (2019).

[S3] L. Wirtz and A. Rubio, The phonon dispersion of graphite revisited, Solid State Commun. 131, 141 (2004).

[S4] A. N. Kolmogorov and V. H. Crespi, Registry-dependent interlayer potential for graphitic systems, Phys. Rev. B 71, 235415 (2005).

[S5] A. A. Maradudin and S. H. Vosko, Symmetry properties of the normal vibrations of a crystal, Rev. Mod. Phys. 40, 1 (1968).

[S6] A. Marek, V. Blum, R. Johanni, V. Havu, B. Lang, T. Auckenthaler, A. Heinecke, H.-J. Bungartz, and H. Lederer, The ELPA Library - Scalable Parallel Eigenvalue Solutions for Electronic Structure Theory and Computational Science, J. Condens. Matter Phys. 26, 213201 (2014).

[S7] T. Nakanishi and T. Ando, Conductance of Crossed Carbon Nanotubes, J. Phys. Soc. Japan 70, 1647 (2001).

[S8] P. Moon and M. Koshino, Energy spectrum and quantum Hall effect in twisted bilayer graphene, Phys. Rev. B 85, 195458 (2012).

[S9] P. E. Blöchl, O. Jepsen, and O. K. Andersen, Improved tetrahedron method for Brillouin-zone integrations, Phys. Rev. B 49, 16223 (1994).