

Strong electron-phonon coupling, electron-hole asymmetry, and nonadiabaticity in magic-angle twisted bilayer graphene

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We report strong electron-phonon coupling in magic-angle twisted bilayer graphene (MA-TBG) obtained from atomistic description of the system including more than 10 000 atoms in the moiré supercell. Electronic structure, phonon spectrum, and electron-phonon coupling strength λ are obtained before and after atomic-position relaxation both in and out of plane. Obtained λ is very large for MA-TBG, with $\lambda > 1$ near the half-filling energies of the flat bands, while it is small ($\lambda \sim 0.1$) for monolayer and unrotated bilayer graphene. Significant electron-hole asymmetry occurs in the electronic structure after atomic-structure relaxation, so λ is much stronger with hole doping than electron doping. Obtained electron-phonon coupling is nearly isotropic and depends very weakly on electronic band and momentum, indicating that electron-phonon coupling prefers single-gap *s*-wave superconductivity. Relevant phonon energies are much larger than electron energy scale, going far beyond adiabatic limit. Our results provide a fundamental understanding of the electron-phonon interaction in MA-TBG, highlighting that it can contribute to rich physics of the system.

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Interplay between the interlayer coupling and the rotational mismatch between two graphene layers in bilayer graphene results in flattening of Dirac cones at certain special twist angles θ_M , called magic angles [1–3]. Recently, correlated insulator behavior and superconductivity are experimentally observed near the first magic angle $\theta_M = 1.08^\circ$, demonstrating rich physics induced by the presence of the flat bands [4,5]. In this regard, more detailed characterizations for the magic-angle twisted bilayer graphene (MA-TBG) are attracting great interest [6–51].

In addition to the exotic electronic properties, it has been observed that low-angle bilayer graphene exhibits atomic-scale reconstruction [52]. The essential effect of the lattice relaxation is such that the area of the AA stacking region becomes smaller, while the AB stacking region is larger, and this effect becomes more important as the twist angle gets smaller. Also, it is suggested that the lattice relaxation can affect the electronic structure, opening superlattice-induced energy gaps at the band edges on both electron and hole sides [53]. Since these gaps are clearly observed in the experiments [4,54], it is necessary to consider the lattice relaxation when studying TBG in the low-angle regime.

As the electron-phonon coupling strength λ in simple monolayer and unrotated bilayer graphene is too weak, superconductivity in MA-TBG is suspected to be originated from the electron correlation. *Ab initio* calculations found that λ of monolayer and unrotated bilayer graphene is less than 0.1 near the charge-neutral Fermi level [55]. If λ has a similar value in MA-TBG, it cannot account for the observed superconducting transition temperature $T_c \sim 1$ K.

However, since λ is proportional to the electron density of states, λ of AB-stacked bilayer graphene (AB-BLG), for

example, can be as large as 0.28 when the Fermi level is tuned to near the van Hove singularity points. This suggests that λ is likely to be further enhanced in low-angle twisted bilayer graphene where the flattening of Dirac cones brings large enhancements of the electron density of states. Thus, quantitative estimation of λ in low-angle twisted bilayer graphene can provide a valuable insight into the nature of superconductivity.

In this work, we investigate the electron-phonon interaction in MA-TBG with atomistic description of the system including more than 10 000 atoms needed for the moiré supercell. We use a tight-binding approach for electrons and atomic force constants for phonons. We find that the electron-phonon coupling strength λ in MA-TBG is almost directly proportional to the electron density of states and becomes greater than 1 near the half-filling energies of the flat bands. It is shown that the lattice relaxations can bring electron-hole asymmetry to the electron density of states and, as a result, the hole-side flat bands have much stronger λ than the electron side. We also find that the electron-phonon coupling depends very weakly on the direction and magnitude of the electronic crystal momentum. We discuss implications of our results for superconductivity in MA-TBG.

Although the electron-phonon interaction can be, in principle, obtained accurately by self-consistent density functional perturbation theory (DFPT), the large number ($\sim 10^4$) of atoms in the moiré supercell is a practical barrier making DFPT calculations very difficult to achieve. In addition, considering correlation effects between electrons in atomistic description also requires challenging development due to the large number of atoms. In our present work, we employ a tight-binding approach with one *p* orbital per carbon atom and atomic force constants for atomic vibrations without considering correlation effects between electrons. Our results provide a fundamental understanding of the electron-phonon

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interaction in the system obtained from atomistic description of electrons and phonons.

A moiré supercell of twisted bilayer graphene is constructed by rotating each layer of AA-stacked bilayer graphene by $\theta/2$ and $-\theta/2$, respectively. The resulting atomic structure has sixfold rotation symmetry axis around the z axis, and three twofold rotation symmetry axes that are perpendicular to the z axis, which swap two graphene layers as a result.

Preserving the crystal symmetry of nonrelaxed structure, we determine the equilibrium atomic positions by minimizing the total energy U that is the sum of in-plane strain energy and interlayer binding energy,

$$U = \frac{1}{2} \sum_{l=1}^2 \sum_{p\kappa\alpha, p'\kappa'\beta} C_{p\kappa\alpha, p'\kappa'\beta}^{\text{MLG}} \Delta\tau_{p\kappa\alpha}^l \Delta\tau_{p'\kappa'\beta}^l + \sum_{p\kappa, p'\kappa'} V_{\text{KC}}(\tau_{p\kappa}^1 - \tau_{p'\kappa'}^2). \quad (1)$$

Here $\tau_{p\kappa\alpha}^l$ is the α ($\alpha = x, y, z$) component of the position of the κ th atom in layer l located at the p th moiré supercell of TBG, $\Delta\tau_{p\kappa}^l = \tau_{p\kappa}^l - \tilde{\tau}_{p\kappa}^l$ is the deviation from the nonrelaxed position $\tilde{\tau}_{p\kappa}^l$, and $C_{p\kappa\alpha, p'\kappa'\beta}^{\text{MLG}}$ are force constants between two atoms in the same layer up to fourth-nearest neighbors, taken from Ref. [56], which are obtained by fitting to the *ab initio* phonon dispersion calculations of monolayer graphene. The interlayer binding energy is calculated using Kolmogorov-Crespi (KC) potential V_{KC} that depends on interlayer atomic registry [57]. Without the interlayer binding energy, our total energy function has its minimum, by construction, at the atomic positions of the rigidly rotated two graphene layers. With the interlayer binding energy, the equilibrium atomic positions show that the area of AA-stacked regions is shrunk, while AB-stacked regions expanded, and interlayer distances in AA-stacked regions become larger than AB-stacked regions [52,53,58,59].

Figure 1 shows the atomic displacements due to the relaxation at $\theta = 1.08^\circ$. We find that maximal out-of-plane displacements are about two times maximal in-plane displacements. Out-of-plane displacements are largest at the AA-stacked region, and also noticeable at the AB/BA domain boundary. The existence of locally confined strains at AB/BA domain boundaries is one of the most important consequences of the lattice relaxations in low-angle TBG. Our results are consistent with previous studies on the lattice relaxations in low-angle TBG.

To investigate electronic structures of TBG in both nonrelaxed and relaxed structure, we employ a single-orbital tight-binding approach where the electronic Hamiltonian is

$$\hat{H} = \sum_{p\kappa, p'\kappa'} t(\tau_{p\kappa} - \tau_{p'\kappa'}) |\phi_\kappa; \mathbf{R}_p\rangle \langle \phi_{\kappa'}; \mathbf{R}_{p'}|, \quad (2)$$

where $|\phi_\kappa; \mathbf{R}_p\rangle$ is a carbon p_z -like orbital at $\tau_{p\kappa}$. Here we drop the layer index on $\tau_{p\kappa}$, κ sweeps all atoms in both layers, and $\tau_{p\kappa} = \tau_{0\kappa} + \mathbf{R}_p$ for the p th moiré supercell at \mathbf{R}_p . We use the Slater-Koster-type hopping integral,

$$t(\mathbf{d}) = V_{pp\pi}^0 e^{-(d-a_0)/\delta} \{1 - (d_z/d)^2\} + V_{pp\sigma}^0 e^{-(d-d_0)/\delta} (d_z/d)^2, \quad (3)$$

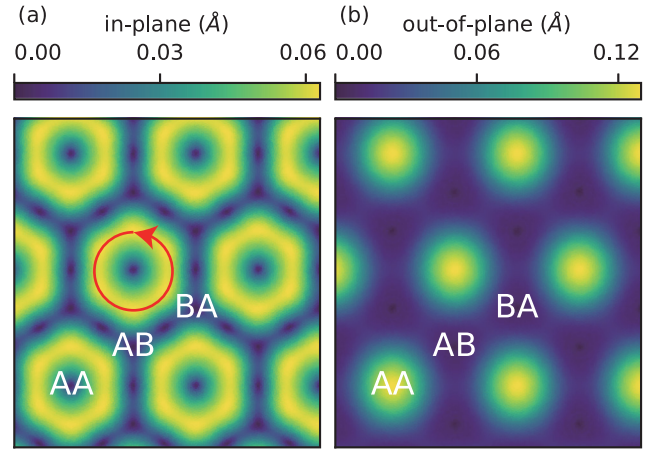


FIG. 1. Magnitude of (a) in-plane and (b) out-of-plane displacements of the upper layer after the structural relaxation in the TBG at $\theta = 1.08^\circ$. Red circular arrow denotes the directions of in-plane displacements. The other layer has a similar displacement pattern, except that the directions are opposite. The stacking pattern of two graphene layers varies within the moiré supercell of TBG. AA-, AB-, and BA-type stacking regions are denoted by AA, AB, and BA, respectively.

where \mathbf{d} is the displacement vector between two orbitals. The hopping energy $V_{pp\pi}^0 = -2.7$ eV is between in-plane nearest neighbors separated by $a_0 = a/\sqrt{3} = 1.42$ Å, and $V_{pp\sigma}^0 = 0.48$ eV is between two vertically aligned atoms at the distance $d_0 = 3.35$ Å. Here $\delta = 0.184a$ is chosen to set the magnitude of the next-nearest-neighbor hopping amplitude to be $0.1V_{pp\pi}^0$ [60,61]. We use the cutoff distance $d_c = 5$ Å, beyond which the hopping integral is negligible.

Figure 2(a) shows the band structures for TBLG at $\theta = 1.08^\circ$ in the nonrelaxed and relaxed structure. One of the most noticeable effects of the lattice relaxation is the opening of the gaps at the edges of the flat bands. Furthermore, the electron-side and hole-side flat bands become significantly asymmetric due to the relaxation. The hole side gets much narrower than the electron side so that the peak height of the density of states [Fig. 2(b)] in the hole side is more than twice the electron side. The gap opening and the electron-hole asymmetry are consistent qualitatively with previous results considering in-plane relaxation only [53].

Figures 2(c) and 2(d) show Fermi surfaces at energies where the hole-side flat bands are half-filled for the nonrelaxed and relaxed structures, respectively. At these energies, Fermi surfaces become more complicated than those near the charge-neutral energy, where only circular Fermi sheets originating from the Dirac cones are located at Brillouin zone corners. At half-filling energies, Fermi sheets at the zone corners become similar to triangles, and the additional Γ -centered Fermi sheets appear.

Phonons in twisted bilayer graphene are calculated using atomic force constants $C_{p\kappa\alpha, p'\kappa'\beta} = \partial^2 U / \partial \tau_{p\kappa\alpha} \partial \tau_{p'\kappa'\beta}$, where U is given by Eq. (1). Since we treat the in-plane strain energy with the harmonic approximation, the in-plane force constants are unaltered by the lattice relaxation. The interlayer force constants, however, are evaluated at relaxed atomic positions

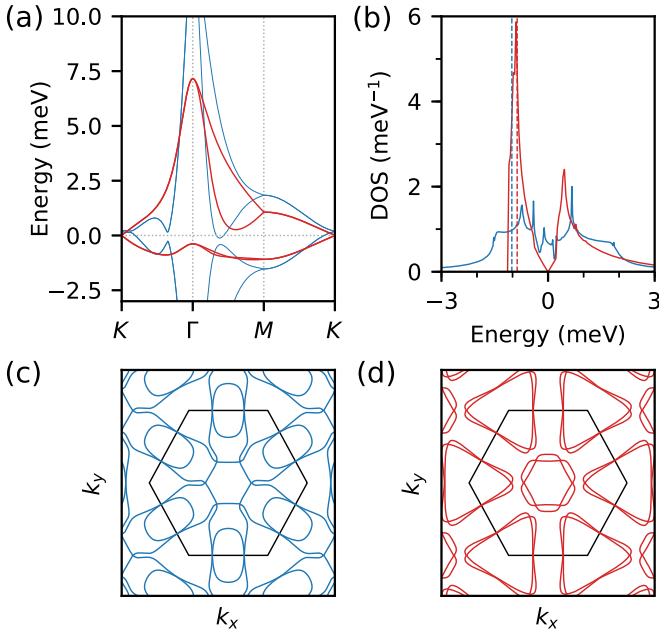


FIG. 2. Electronic structure of MA-TBG. Tight-binding (a) band structure and (b) density of states per spin per moiré supercell for nonrelaxed (blue) and relaxed (red) structures at $\theta = 1.08^\circ$. Vertical dashed lines show hole-side half-filling energies. (c),(d) Fermi surfaces at energies denoted by dashed lines in (b) for nonrelaxed and relaxed structures, respectively.

because the KC potential [57] is not harmonic. Our approach is similar to Ref. [62], except that the Lennard-Jones interlayer potential between two graphene layers is replaced by the KC potential, which can account for registry-dependent energy differences in TBG. From the force constants, we obtain the dynamical matrix $D_{\kappa\alpha,\kappa'\beta}(\mathbf{q}) = \sum_p e^{i\mathbf{q}\cdot\mathbf{R}_p} C_{0\kappa\alpha,p\kappa'\beta} / M_C$ for phonon wave vector \mathbf{q} , where M_C is the mass of a carbon atom. Then, we solve the phonon eigenvalue problems $\omega_{qv}^2 e_{qv,\kappa\alpha} = \sum_{\kappa',\beta} D_{\kappa\alpha,\kappa'\beta}(\mathbf{q}) e_{qv,\kappa'\beta}$ at the irreducible Brillouin zone of TBG for the energy ω_{qv} and polarization vector $e_{qv,\kappa}$ of the v th phonon mode. The phonons in the rest of the Brillouin zone are obtained from the symmetry relations [63]. We considered all phonon modes in the moiré supercell to obtain unbiased results for electron-phonon interaction.

Figure 3(a) shows phonon density of states $F(\omega)$ for $\theta = 1.08^\circ$, 1.12° , and 1.16° as well as AB-BLG. Phonon spectra are nearly insensitive to small twist-angle differences. So a tiny difference is that, compared to AB-BLG, interlayer breathing modes near $\omega \sim 11$ meV are slightly softened in TBG [see Fig. S1(a) in the Supplemental Material [64] for phonon dispersions in AB-BLG].

Now, we calculate the standard electron-phonon coupling strengths defined as

$$\lambda_{nk} = 2N_F \sum_{mqv} \frac{|g_{mnv}(\mathbf{k}, \mathbf{q})|^2}{\omega_{qv}} W_{m\mathbf{k}+\mathbf{q}}, \quad (4a)$$

$$\lambda = \sum_{nk} \lambda_{nk} W_{nk}, \quad (4b)$$

where N_F is the electron density of states per spin at the Fermi level E_F , and $W_{nk} = \delta(E_F - \varepsilon_{nk}) / N_F$ is the partial

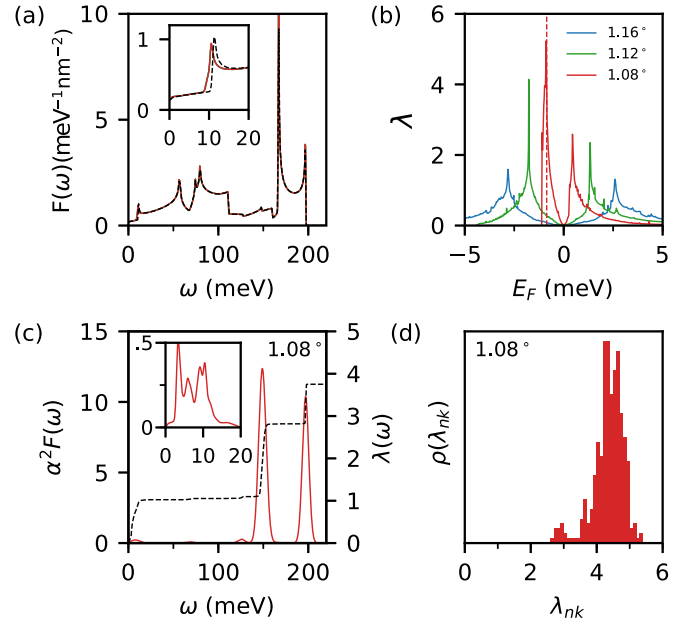


FIG. 3. (a) Phonon density of states for AB-BLG (dashed black), TBG at $\theta = 1.08^\circ$ (solid red), $\theta = 1.12^\circ$ (solid green), and $\theta = 1.16^\circ$ (solid blue). Phonons are insensitive to the small twist-angle differences between those angles. The inset shows the frequency range of the interlayer shear and breathing modes, which are softened by the twist. (b) Total electron-phonon coupling strength λ in TBG as a function of the Fermi energy (E_F). The vertical red dashed line denotes the energy where hole-side flat bands in $\theta = 1.08^\circ$ are half-filled. (c) Eliashberg function $\alpha^2 F(\omega)$, shown in red, at the half-filling energy in the hole side. The dashed black line denotes $\lambda(\omega) = 2 \int_0^\omega \alpha^2 F(\omega') / \omega' d\omega'$. The inset shows the low-frequency range of $\alpha^2 F(\omega)$. Phonon modes at this range contribute to about 30% of the total coupling strength. (d) Distribution of band- and momentum-resolved coupling strength λ_{nk} of Eq. (4a) at the hole-side half-filling energy.

weight of the density of states. Here, ε_{nk} is the electron energy of the n th band with wave vector \mathbf{k} , and W_{nk} is obtained by the linear tetrahedron method [65]. The electron-phonon matrix elements $g_{mnv}(\mathbf{k}, \mathbf{q}) = \langle m\mathbf{k} + \mathbf{q} | \delta_{qv} \hat{H} | n\mathbf{k} \rangle$ couple the electronic states $|n\mathbf{k}\rangle$ and $|m\mathbf{k} + \mathbf{q}\rangle$, where $\delta_{qv} \hat{H}$ is the change in \hat{H} due to phonon mode (qv). The electron-phonon matrix elements in localized orbital basis can be expressed in terms of the changes in the hopping matrix elements due to the atomic displacements of phonon modes [66–68],

$$g_{mnv}(\mathbf{k}, \mathbf{q}) = l_{qv} \sum_{\kappa\alpha} e_{qv,\kappa\alpha} \sum_{pp',ij} e^{-i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}_{p'}} e^{i\mathbf{k}\cdot\mathbf{R}_p} \times c_{m\mathbf{k}+\mathbf{q},j}^* c_{n\mathbf{k},i} \langle \phi_j; \mathbf{R}_{p'} | \frac{\partial \hat{H}}{\partial \tau_{0\kappa\alpha}} | \phi_i; \mathbf{R}_p \rangle, \quad (5)$$

where $l_{qv} = \sqrt{\hbar / (2M_C \omega_{qv})}$ is the length scale of phonon mode (qv), and $c_{n\mathbf{k},i}$ is the coefficient of the electron wave functions in local orbital basis, i.e., $c_{n\mathbf{k},i} e^{i\mathbf{k}\cdot\mathbf{R}_p} = \sqrt{N} \langle \phi_i; \mathbf{R}_p | n\mathbf{k} \rangle$. Here, N is the total number of unit cells over which the electronic states are normalized. Thus, in our tight-binding approach, we obtain the electron-phonon matrix

elements

$$g_{mn\nu}(\mathbf{k}, \mathbf{q}) = l_{q\nu} \sum_{\kappa\alpha} e_{q\nu,\kappa\alpha} \sum_{p,i} \frac{\partial}{\partial x_\alpha} t(\boldsymbol{\tau}_{0\kappa} - \boldsymbol{\tau}_{pi}) \times \{e^{i\mathbf{k}\cdot\mathbf{R}_p} c_{m\mathbf{k}+\mathbf{q},\kappa}^* c_{n\mathbf{k},i} + e^{-i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}_p} c_{m\mathbf{k}+\mathbf{q},i}^* c_{n\mathbf{k},\kappa}\}. \quad (6)$$

When we apply our method to calculate λ for simple monolayer graphene and AB-BLG, λ is less than 0.1 near the charge-neutral energy but it increases up to 0.2–0.3 in proportion to the density of states when the chemical potential is varied [Fig. S1(b) [64]]. This is consistent with the previous studies for monolayer and bilayer graphene [55,69,70].

Figure 3(b) shows calculated electron-phonon coupling strength as a function of the Fermi energy (E_F) for the three twist angles. \mathbf{k} and \mathbf{q} grids of 30×30 in the moiré Brillouin zone are used for electrons and phonons. Due to the large density of states of the flat bands, λ becomes extremely large as θ approaches 1.08° , where the Dirac cones are nearly flat. The average interaction between electronic states, λ/N_F , for $\theta = 1.08^\circ$ is approximately twice that for $\theta = 0$. Furthermore, as the lattice relaxation brings electron-hole asymmetry in the density of states, the maximum value of λ in the hole-side flat bands is almost twice that in the electron side for $\theta = 1.08^\circ$.

Figure 3(c) shows the isotropic Eliashberg function $\alpha^2 F(\omega) = \frac{1}{N_F} \sum_{nm\nu\mathbf{k}\mathbf{q}} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \delta(E_F - \varepsilon_{n\mathbf{k}}) \delta(E_F - \varepsilon_{m\mathbf{k}+\mathbf{q}}) \delta(\omega - \omega_{q\nu})$ at the half-filling energy of the hole-side flat bands in $\theta = 1.08^\circ$. With $\alpha^2 F(\omega)$, λ of Eq. (4b) is equal to $\lambda = 2 \int_0^\infty \alpha^2 F(\omega) / \omega d\omega$. In Fig. 3(c), in-plane optical modes generate strong peaks at 150 and 200 meV in $\alpha^2 F(\omega)$, contributing to about 70% of λ . Although the interlayer shear (~ 2 meV) and breathing modes (~ 11 meV) have an order of magnitude smaller values of $\alpha^2 F(\omega)$ than the in-plane optical modes, they have significant contributions to λ due to their low-phonon energies. We also find $\lambda_{n\mathbf{k}}$ is nearly isotropic and depends very weakly on the electronic band and momentum [Fig. 3(d)], which indicates the electron-phonon coupling prefers single-gap s -wave superconductivity [71].

In conventional phonon-mediated superconductors, transition temperature can be reliably calculated from the Migdal-Eliashberg equations [72–74]. But the validity of the Migdal-Eliashberg theory depends on the existence of small parameter $\omega_{\text{ph}}/E_F \ll 1$ where ω_{ph} is relevant phonon energy scale. This condition is obviously violated in magic-angle twisted bilayer graphene. For instance, while $E_F \approx 1$ meV near the half-fillings of the flat bands, $\omega_{\text{ph}} \approx 2 \sim 11$ meV for the interlayer shear and breathing modes, and $\omega_{\text{ph}} \approx 150 \sim 200$ meV for

the in-plane optical modes. In this sense, MA-TBG systems are close to the adiabatic limit $\omega_{\text{ph}}/E_F \gg 1$.

In the adiabatic limit, T_c was studied in several literatures [75–79], where the prefactor of T_c is determined by E_F instead of the phonon energy [77,79], that is,

$$T_c \sim E_F \exp(-1/\lambda). \quad (7)$$

In our calculations for $\theta = 1.08^\circ$, $\lambda = 3.6$ (0.56) at $E_F = 0.86$ (1.02) meV when the hole-side (electron-side) flat bands are half-filled. These values give $T_c \sim 7.5$ K for the hole side and $T_c \sim 1.9$ K for the electron side. Although our estimation is crude for direct comparison with experiments, the order of magnitude is close to the experimentally observed $T_c \sim 1.7$ K in the hole side. Since our estimation did not consider the effect of Coulomb interaction, which can reduce T_c , we expect that calculating T_c including the Coulomb effect can give more consistent results to the experimental situations. Also, the rapid energy dependence of the electronic density of states can play an important role in determining T_c [80].

In conclusion, we have calculated the electron-phonon coupling strength in the magic-angle twisted bilayer graphene using atomistic description of electrons and phonons. Obtained λ in MA-TBG becomes almost an order of magnitude larger than that in simple monolayer or unrotated bilayer graphene. For $\theta = 1.08^\circ$, the electron-hole asymmetry arises from atomic-structure relaxation due to interlayer interaction so that the electron-phonon coupling is stronger in the hole-side flat bands. The obtained electron-phonon interaction is almost isotropic and depends very weakly on the electronic band and momentum, which indicates the electron-phonon coupling prefers single-gap s -wave superconductivity. We also found that MA-TBG is in the adiabatic limit where the electron energy scale is much smaller than the phonon energy scale. Although the T_c formula in the adiabatic limit produces values of T_c comparable to the experiments, theory of T_c of the system may require including Coulomb interaction and rapid energy dependence of the electronic density of states as well as electron correlation and any possible presence of magnetic fluctuations. Our results provide a fundamental understanding of the electron-phonon interaction in MA-TBG obtained from an atomistic description of electrons and phonons, highlighting that it can contribute to rich physics of the system.

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- [1] T. de Laissardi re, D. Mayou, and L. Magaud, Localization of Dirac electrons in rotated graphene bilayers, *Nano Lett.* **10**, 804 (2010).
- [2] E. S. Morell, J. D. Correa, P. Vargas, M. Pacheco, and Z. Barticevic, Flat bands in slightly twisted bilayer graphene: Tight-binding calculations, *Phys. Rev. B* **82**, 121407 (2010).
- [3] R. Bistritzer and A. MacDonald, Moir  bands in twisted double-layer graphene, *Proc. Natl. Acad. Sci. USA* **108**, 12233 (2011).

- [4] Y. Cao, V. Fatemi, A. Demir, S. Fang, S. L. Tomarken, J. Y. Luo, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras, R. C. Ashoori, and P. Jarillo-Herrero, Correlated insulator behavior at half-filling in magic-angle graphene superlattices, *Nature (London)* **556**, 80 (2018).
- [5] 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).

- [6] H. Guo, X. Zhu, S. Feng, and R. T. Scalettar, Pairing symmetry of interacting fermions on a twisted bilayer graphene superlattice, *Phys. Rev. B* **97**, 235453 (2018).
- [7] J. F. Dodaro, S. A. Kivelson, Y. Schattner, X. Q. Sun, and C. Wang, Phases of a phenomenological model of twisted bilayer graphene, *Phys. Rev. B* **98**, 075154 (2018).
- [8] M. Fidrysiak, M. Zegrodnik, and J. Spałek, Unconventional topological superconductivity and phase diagram for an effective two-orbital model as applied to twisted bilayer graphene, *Phys. Rev. B* **98**, 085436 (2018).
- [9] T.-F. Chung, Y. Xu, and Y. P. Chen, Transport measurements in twisted bilayer graphene: Electron-phonon coupling and Landau level crossing, *Phys. Rev. B* **98**, 035425 (2018).
- [10] M. Ochi, M. Koshino, and K. Kuroki, Possible correlated insulating states in magic-angle twisted bilayer graphene under strongly competing interactions, *Phys. Rev. B* **98**, 081102 (2018).
- [11] A. Thomson, S. Chatterjee, S. Sachdev, and M. S. Scheurer, Triangular antiferromagnetism on the honeycomb lattice of twisted bilayer graphene, *Phys. Rev. B* **98**, 075109 (2018).
- [12] L. Zou, H. C. Po, A. Vishwanath, and T. Senthil, Band structure of twisted bilayer graphene: Emergent symmetries, commensurate approximants, and Wannier obstructions, *Phys. Rev. B* **98**, 085435 (2018).
- [13] X. Lin and D. Tománek, Minimum model for the electronic structure of twisted bilayer graphene and related structures, *Phys. Rev. B* **98**, 081410 (2018).
- [14] S. Carr, S. Fang, P. Jarillo-Herrero, and E. Kaxiras, Pressure dependence of the magic twist angle in graphene superlattices, *Phys. Rev. B* **98**, 085144 (2018).
- [15] M. Anđelković, L. Covaci, and F. M. Peeters, DC conductivity of twisted bilayer graphene: Angle-dependent transport properties and effects of disorder, *Phys. Rev. Mater.* **2**, 034004 (2018).
- [16] H. Chun Po, L. Zou, A. Vishwanath, and T. Senthil, Origin of Mott Insulating behavior and Superconductivity in Twisted Bilayer Graphene, *Phys. Rev. X* **8**, 031089 (2018).
- [17] B. Roy and V. Juricic, Unconventional superconductivity in nearly flat bands in twisted bilayer graphene, [arXiv:1803.11190](https://arxiv.org/abs/1803.11190).
- [18] G. Baskara, Theory of emergent Josephson lattice in neutral twisted bilayer graphene (Moiré is different), [arXiv:1804.00627](https://arxiv.org/abs/1804.00627).
- [19] B. Padhi, C. Setty, and P. W. Phillips, Doped twisted bilayer graphene near magic angles: Proximity to Wigner crystallization not Mott insulation, *Nano Lett.* **18**, 6175 (2018).
- [20] T. Huang, L. Zhang, and T. Ma, Antiferromagnetically ordered Mott insulator and $d + id$ superconductivity in twisted bilayer graphene: A quantum Monte Carlo study, [arXiv:1804.06096](https://arxiv.org/abs/1804.06096).
- [21] L. Zhang, Low-energy Moiré band formed by Dirac zero modes in twisted bilayer graphene, [arXiv:1804.09047](https://arxiv.org/abs/1804.09047).
- [22] S. Ray and T. Das, Wannier pairs in the superconducting twisted bilayer graphene and related systems, [arXiv:1804.09674](https://arxiv.org/abs/1804.09674).
- [23] C.-C. Liu, L.-D. Zhang, W.-Q. Chen, and F. Yang, Chiral SDW and $d + id$ Superconductivity in the Magic-Angle-Twisted Bilayer Graphene, *Phys. Rev. Lett.* **121**, 217001 (2018).
- [24] X. Y. Xu, K. T. Law, and P. A. Lee, Kekulé valence order in an extended Hubbard model on the honeycomb lattice with possible applications to twisted bilayer graphene, *Phys. Rev. B* **98**, 121406 (2018).
- [25] T. J. Peltonen, R. Ojajarvi, and T. T. Heikkilä, Mean-field theory for superconductivity in twisted bilayer graphene, *Phys. Rev. B* **98**, 220504(R) (2018).
- [26] J.-B. Qiao, L.-J. Yin, and L. He, Twisted graphene bilayer around the first magic angle engineered by heterostrain, *Phys. Rev. B* **98**, 235402 (2018).
- [27] J. Kang and O. Vafek, Symmetry, Maximally Localized Wannier States, and Low Energy Model for the Twisted Bilayer Graphene Narrow Bands, *Phys. Rev. X* **8**, 031088 (2018).
- [28] L. Rademaker and P. Mellado, Charge-transfer insulation in twisted bilayer graphene, [arXiv:1805.05294](https://arxiv.org/abs/1805.05294).
- [29] D. M. Kennes, J. Lischner, and C. Karrasch, Strong correlations and $d + id$ superconductivity in twisted bilayer graphene, *Phys. Rev. B* **98**, 241407(R) (2018).
- [30] H. Isobe, N. F. Q. Yuan, and L. Fu, Unconventional Superconductivity and Density Waves in Twisted Bilayer Graphene, *Phys. Rev. X* **8**, 041041 (2018).
- [31] M. Koshino, N. F. Q. Yuan, T. Koretsune, M. Ochi, K. Kuroki, and L. Fu, Maximally-Localized Wannier Orbitals and the Extended Hubbard Model for the Twisted Bilayer Graphene, *Phys. Rev. X* **8**, 031087 (2018).
- [32] Y.-Z. You and A. Vishwanath, Superconductivity from valley fluctuations and approximate SO(4) symmetry in a weak coupling theory of twisted bilayer graphene, [arXiv:1805.06867](https://arxiv.org/abs/1805.06867).
- [33] J. M. Pizarro, M. J. Calderón, and E. Bascones, The nature of correlations in the insulating states of twisted bilayer graphene, [arXiv:1805.07303](https://arxiv.org/abs/1805.07303).
- [34] F. Wu, A. H. MacDonald, and I. Martin, Theory of phonon-mediated superconductivity in twisted bilayer graphene, *Phys. Rev. Lett.* **121**, 257001 (2018).
- [35] H. K. Pal, On magic angles and band flattening in twisted bilayer graphene, [arXiv:1805.08803](https://arxiv.org/abs/1805.08803).
- [36] F. Guinea and N. R. Walet, Electrostatic effects, band distortions, and superconductivity in twisted graphene bilayers, *PNAS* (2018), doi: [10.1073/pnas.1810947115](https://doi.org/10.1073/pnas.1810947115).
- [37] J. Gonzalez and T. Staube, Kohn-Luttinger superconductivity in twisted bilayer graphene, [arXiv:1807.01275](https://arxiv.org/abs/1807.01275).
- [38] Y. Su and S.-Z. Li, Spontaneous vortex-antivortex lattice in superconducting twisted bilayer graphene, *Phys. Rev. B* **98**, 195101 (2018).
- [39] B. Lian, Z. Wang, and B. A. Bernevig, Twisted bilayer graphene: A phonon driven superconductor, [arXiv:1807.04382](https://arxiv.org/abs/1807.04382).
- [40] Y. Sherkunov and J. J. Betouras, Novel phases in twisted bilayer graphene at magic angles as a result of van Hove singularities and interactions, *Phys. Rev. B* **98**, 205151 (2018).
- [41] A. O. Sboychakov, A. V. Rozhkov, A. L. Rakhmanov, and F. Nori, Many-body effects in twisted bilayer graphene at low twist angles, [arXiv:1807.08190](https://arxiv.org/abs/1807.08190).
- [42] B. L. Chittari, N. Leconte, S. Javvaji, and J. Jung, Pressure induced compression of flatbands in twisted bilayer graphene, [arXiv:1808.00104](https://arxiv.org/abs/1808.00104).
- [43] K. Hejazi, C. Liu, H. Shapourian, X. Chen, and L. Balents, Multiple topological transitions in twisted bilayer graphene near the first magic angle, [arXiv:1808.01568](https://arxiv.org/abs/1808.01568).
- [44] E. Laksono, J. N. Leaw, A. Reaves, M. Singh, X. Wang, S. Adam, and X. Gu, Singlet superconductivity enhanced by charge order in nested twisted bilayer graphene Fermi surfaces, *Solid State Commun.* **282**, 38 (2018).

- [45] G. Tarnopolsky, A. J. Kruchkov, and A. Vishwanath, Origin of magic angles in twisted bilayer graphene, [arXiv:1808.05250](https://arxiv.org/abs/1808.05250).
- [46] J. Ahn, S. Park, and B.-J. Yang, Failure of Nielsen-Ninomiya theorem and fragile topology in two-dimensional systems with space-time inversion symmetry: Application to twisted bilayer graphene at magic angle, [arXiv:1808.05375](https://arxiv.org/abs/1808.05375).
- [47] M. Yankowitz, S. Chen, H. Polshyn, K. Watanabe, T. Taniguchi, D. Graf, A. F. Young, and C. R. Dean, Tuning superconductivity in twisted bilayer graphene, [arXiv:1808.07865](https://arxiv.org/abs/1808.07865).
- [48] J. W. F. Venderbos and R. M. Fernandes, Correlations and electronic order in a two-orbital honeycomb lattice model for twisted bilayer graphene, *Phys. Rev. B* **98**, 245103 (2018).
- [49] L. Chen, H.-Z. Li, and R.-S. Han, Magnetic-impurity resonance states for different pairing symmetries in twisted bilayer graphene, [arXiv:1809.00436](https://arxiv.org/abs/1809.00436).
- [50] T. Stauber, T. Low, and G. Gómez-Santo, Linear response of twisted bilayer graphene: Continuum versus tight-binding models, *Phys. Rev. B* **98**, 195414 (2018).
- [51] Q. K. Tang, L. Yang, D. Wang, F. C. Zhang, and Q. H. Wang, Spin-triplet f -wave pairing in twisted bilayer graphene near $1/4$ filling, [arXiv:1809.06772](https://arxiv.org/abs/1809.06772).
- [52] H. Yoo, K. Zhang, R. Engelke, P. Cazeaux, S. H. Sung, R. Hovden, A. W. Tsen, T. Taniguchi, K. Watanabe, G.-C. Yi, M. Kim, M. Lusk, E. B. Tadmor, and P. Kim, Atomic reconstruction at van der Waals interface in twisted bilayer graphene, [arXiv:1804.03806](https://arxiv.org/abs/1804.03806).
- [53] N. N. T. Nam and M. Koshino, Lattice relaxation and energy band modulation in twisted bilayer graphene, *Phys. Rev. B* **96**, 075311 (2017).
- [54] Y. Cao, J. Y. Luo, V. Fatemi, S. Fang, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, Superlattice-Induced Insulating States and Valley-Protected Orbits in Twisted Bilayer Graphene, *Phys. Rev. Lett.* **117**, 116804 (2016).
- [55] C.-H. Park, F. Giustino, M. L. Cohen, and S. G. Louie, Electron-Phonon Interactions in Graphene, Bilayer Graphene, and Graphite, *Nano Lett.* **8**, 4229 (2008).
- [56] L. Wirtz and A. Rubio, The phonon dispersion of graphite revisited, *Solid State Commun.* **131**, 141 (2004).
- [57] A. N. Kolmogorov and V. H. Crespi, Registry-dependent interlayer potential for graphitic systems, *Phys. Rev. B* **71**, 235415 (2005).
- [58] K. Zhang and E. B. Tadmor, Energy and Moiré patterns in 2D bilayers in translation and rotation: A study using an efficient discrete-continuum interlayer potential, *Extreme Mech. Lett.* **14**, 16 (2017).
- [59] K. Zhang and E. B. Tadmor, Structural and electron diffraction scaling of twisted graphene bilayers, *J. Mech. Phys. Solids* **112**, 225 (2018).
- [60] P. Moon and M. Koshino, Energy spectrum and quantum Hall effect in twisted bilayer graphene, *Phys. Rev. B* **85**, 195458 (2012).
- [61] T. Nakanishi and T. Ando, Conductance of crossed carbon nanotubes, *J. Phys. Soc. Jpn.* **70**, 1647 (2001).
- [62] A. Cocemasov, D. Nika, and A. Balandin, Phonons in twisted bilayer graphene, *Phys. Rev. B* **88**, 035428 (2013).
- [63] A. A. Maradudin and S. H. Vosko, Symmetry properties of the normal vibrations of a crystal, *Rev. Mod. Phys.* **40**, 1 (1968).
- [64] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.98.241412> for phonon dispersions of AB-stacked bilayer graphene and electron-phonon coupling strengths for monolayer graphene and AB-stacked bilayer graphene.
- [65] P. E. Blöchl, O. Jepsen, and O. K. Andersen, Improved tetrahedron method for Brillouin-zone integrations, *Phys. Rev. B* **49**, 16223 (1994).
- [66] F. Giustino, M. L. Cohen, and S. G. Louie, Electron-phonon interaction using Wannier functions, *Phys. Rev. B* **76**, 165108 (2007).
- [67] T. Gunst, T. Markussen, K. Stokbro, and M. Brandbyge, First-principles method for electron-phonon coupling and electron mobility: Applications to two-dimensional materials, *Phys. Rev. B* **93**, 035414 (2016).
- [68] L. A. Agapito and M. Bernadi, *Ab initio* electron-phonon interactions using atomic orbital wave functions, *Phys. Rev. B* **97**, 235146 (2018).
- [69] M. Bianchi, E. D. L. Rienks, S. Lizzit, A. Baraldi, R. Balog, L. Hornekær, and P. Hofmann, Electron-phonon coupling in potassium-doped graphene: Angle-resolved photoemission spectroscopy, *Phys. Rev. B* **81**, 041403 (2010).
- [70] J. C. Johannsen, S. Ulstrup, M. Bianchi, R. Hatch, D. Guan, F. Mazzola, L. Hornekær, F. Fromm, C. Roidel, T. Seyller, and P. Hofmann, Electron-phonon coupling in quasi-free-standing graphene, *J. Phys.: Condens. Matter* **25**, 094001 (2013).
- [71] If distribution of λ_{nk} formed multiple groups in Fig. 3(d), it would indicate multiple-gap s -wave superconductivity as in MgB₂. However, in Fig. 3(d), distribution of λ_{nk} forms a single group centered at 4.2. This indicates that the electron-phonon interaction in MA-TBG prefers a single uniform superconducting energy gap.
- [72] A. B. Migdal, Interaction between electrons and lattice vibrations in a normal metal, *Sov. Phys. JETP* **7**, 996 (1958).
- [73] G. M. Eliashberg, Interactions between electrons and lattice vibrations in a superconductor, *Sov. Phys. JETP* **11**, 696 (1960).
- [74] P. B. Allen and R. C. Dynes, Transition temperature of strongly coupled superconductors reanalyzed, *Phys. Rev. B* **12**, 905 (1975).
- [75] D. M. Eagles, Predicted Transition Temperatures of Very Thin Films and Whiskers of Superconducting Semiconductors—Application to SrTiO₃, *Phys. Rev.* **164**, 489 (1967).
- [76] M. A. Ikeda, A. Ogasawara, and M. Sugihara, On Migdal's theorem, *Phys. Lett. A* **170**, 319 (1992).
- [77] L. P. Gor'Kov, Superconducting transition temperature: Interacting Fermi gas and phonon mechanisms in the nonadiabatic regime, *Phys. Rev. B* **93**, 054517 (2016).
- [78] L. P. Gor'Kov, Phonon mechanism in the most dilute superconductor n -type SrTiO₃, *Proc. Natl. Acad. Sci. USA* **113**, 4646 (2016).
- [79] M. V. Sadoskii, Electron-phonon coupling in Eliashberg-McMillan theory beyond adiabatic approximation, [arXiv:1809.02531](https://arxiv.org/abs/1809.02531). Here, the T_c formula is derived for a phonon spectrum which can be represented by a single characteristic frequency. Thus our use of the T_c formula is approximate in the sense that the phonon spectrum of TBG from 2 to 200 meV is represented by a single characteristic frequency although the frequency does not appear in the T_c formula.
- [80] P. B. Allen and B. Mitrović, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1982), Vol. 37, p. 1.