Interacting Twisted Bilayer Graphene with Systematic Modeling of Structural Relaxation

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Modeling Structural Relaxation

To minimize total mechanical energy, the atoms in TBG are displaced depending on the local disregistry. The displacement $\mathbf{U}_\ell$ moves atoms on layer $\ell \in \{1,2\}$, sublattice $\sigma \in \{\mathrm{A},\mathrm{B}\}$ by

$$\mathbf{R}_\ell + \boldsymbol{\tau}_\ell^\sigma \mapsto \mathbf{R}_\ell + \boldsymbol{\tau}_\ell^\sigma + \mathbf{U}_\ell(\mathbf{R}_\ell + \boldsymbol{\tau}_\ell^\sigma)$$

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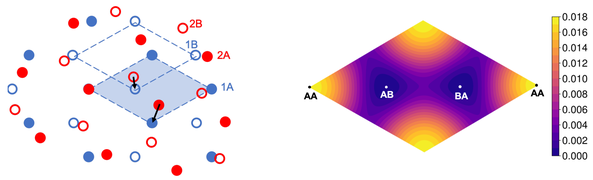


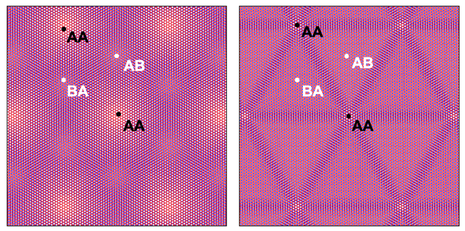
Left: Illustration of disregistry of layer 2 (red) with respect to layer 1 (blue). Right: GSFE energy landscape as a function of disregistry. It is minimized at AB configuration, and maximized at AA.

$\mathbf{U}_\ell$ minimizes the bilayer energy functional $\mathcal{E}[\mathbf{U}_1,\mathbf{U}_2] := \sum_{\ell=1}^{2} \mathcal{E}_\ell^{\text{intra}}(\mathbf{U}_\ell) + \mathcal{E}^{\text{inter}}(\mathbf{U}_1 - \mathbf{U}_2)$ [1].

$$\mathcal{E}_\ell^{\text{intra}}(\mathbf{U}) := \int_{\Gamma_{\mathrm{m}}} \frac{\lambda}{2}(\operatorname{div}\mathbf{U})^2 + \mu\varepsilon(\mathbf{U}) : \varepsilon(\mathbf{U})\,\mathrm{d}\mathbf{x}, \quad \varepsilon(\mathbf{U}) := \frac{1}{2}\left(\nabla\mathbf{U} + \nabla\mathbf{U}^\top\right),$$

$$\mathcal{E}^{\text{inter}}(\mathbf{V}) := \frac{1}{2}\int_{\Gamma_{\mathrm{m}}} \Phi_1^{\text{GSFE}}(\boldsymbol{\gamma}_2(\mathbf{x}) + \mathbf{V}(\mathbf{x})) + \Phi_2^{\text{GSFE}}(\boldsymbol{\gamma}_1(\mathbf{x}) - \mathbf{V}(\mathbf{x}))\,\mathrm{d}\mathbf{x},$$

where $\boldsymbol{\gamma}_2(\mathbf{x}) = (I - A_2A_1^{-1})\mathbf{x}$ is the local stacking configuration of layer 1 with respect to layer 2.



Original Relaxed

Single Particle Hamiltonian

The tight-binding Hamiltonian acts on wave functions through

$$(H\Psi)^\sigma_{\mathbf{R}_1} = \sum_{\mathbf{R}_1'\in\mathcal{R}_1}\sum_{\sigma'} h_{11}(\mathbf{R}_1 + \boldsymbol{\tau}_1^\sigma + \mathbf{U}_1(\mathbf{R}_1 + \boldsymbol{\tau}_1^\sigma) - \mathbf{R}_1' - \boldsymbol{\tau}_1^{\sigma'} - \mathbf{U}_1(\mathbf{R}_1' + \boldsymbol{\tau}_1^{\sigma'}))\Psi^{\sigma'}_{\mathbf{R}_1'}$$
$$+ \sum_{\mathbf{R}_2'\in\mathcal{R}_2}\sum_{\sigma'} h_{12}(\mathbf{R}_1 + \boldsymbol{\tau}_1^\sigma + \mathbf{U}_1(\mathbf{R}_1 + \boldsymbol{\tau}_1^\sigma) - \mathbf{R}_2 - \boldsymbol{\tau}_2^{\sigma'} - \mathbf{U}_2(\mathbf{R}_2 + \boldsymbol{\tau}_2^{\sigma'}))\Psi^{\sigma'}_{\mathbf{R}_2}$$

h_{11} and h_{12} are the intralayer and interlayer hopping functions based on DFT calculations [2]. A multiple scales expansion of the tight-binding model gives effective Hamiltonians:

Bistritzer-MacDonald Model [3] Set $\mathbf{U}_\ell = 0$ and keep first order terms [4].

$$[H_{\mathrm{BM}}(\mathbf{k})]_{\mathbf{G},\mathbf{G}'} = \begin{pmatrix} v\boldsymbol{\sigma}\cdot\left(\mathbf{k} + \frac{\mathbf{s}_1}{2} + \mathbf{G}\right)\delta_{\mathbf{G},\mathbf{G}'} & \sum_{j=0}^{2} T_j\delta_{\mathbf{G},\mathbf{G}'+\mathbf{Q}_j} \\ \sum_{j=0}^{2} T_j^\dagger\delta_{\mathbf{G},\mathbf{G}'-\mathbf{Q}_j} & v\boldsymbol{\sigma}\cdot\left(\mathbf{k} - \frac{\mathbf{s}_1}{2} + \mathbf{G}\right)\delta_{\mathbf{G},\mathbf{G}'} \end{pmatrix} \tag{1}$$

Relaxed BM Model Include higher-order [5] and relaxation corrections.

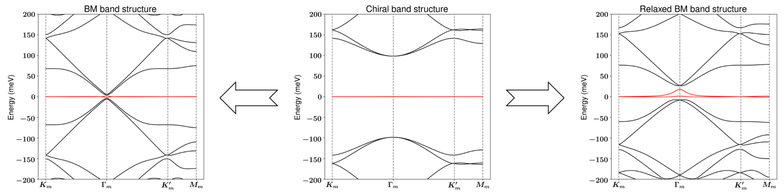
$$[H_{\mathrm{relax}}(\mathbf{k})]_{\mathbf{G},\mathbf{G}'} = \begin{pmatrix} D^{(1)}\left(\mathbf{k} + \frac{\mathbf{s}_1}{2} + \mathbf{G}\right)\delta_{\mathbf{G},\mathbf{G}'} + \sum_{j=1}^{12} A_j^{(1)}\left(\mathbf{k} + \frac{\mathbf{s}_1}{2}\right)\delta_{\mathbf{G},\mathbf{G}'+\mathbf{P}_j} & \sum_{j=1}^{12}\tilde{T}_j(\mathbf{k})\delta_{\mathbf{G},\mathbf{G}'+\mathbf{Q}_j} \\ \sum_{j=1}^{12}\tilde{T}_j^\dagger(\mathbf{k})\delta_{\mathbf{G},\mathbf{G}'-\mathbf{Q}_j} & D^{(2)}\left(\mathbf{k} - \frac{\mathbf{s}_1}{2} + \mathbf{G}\right)\delta_{\mathbf{G},\mathbf{G}'} + \sum_{j=1}^{12} A_j^{(2)}\left(\mathbf{k} - \frac{\mathbf{s}_1}{2}\right)\delta_{\mathbf{G},\mathbf{G}'+\mathbf{P}_j} \end{pmatrix} \tag{2}$$

Intra and interlayer scattering terms are of the form $A_j^{(\ell)}(\mathbf{k}) = A_j^{(\ell)} + A_{j,\nabla}^{(\ell)}\cdot\mathbf{k}$, $\quad \tilde{T}_j(\mathbf{k}) = \tilde{T}_j + \tilde{T}_{j,\nabla}\cdot\mathbf{k}$.

Numerical Simulation

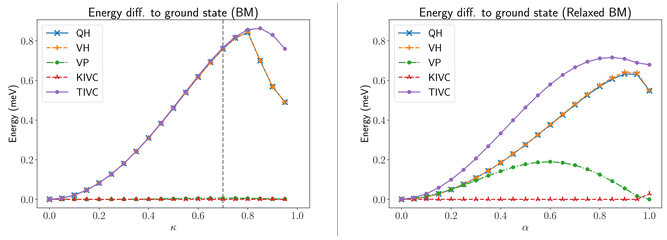
Numerical Setup

Families of Hamiltonians that interpolate between the chiral limit and BM or relaxed BM models $H_0^{\mathrm{BM}}(\kappa) = (1-\kappa)H_{\mathrm{chiral}} + \kappa H_{\mathrm{BM}}$, $H_0^{\mathrm{relaxed}}(\alpha) = (1-\alpha)H_{\mathrm{chiral}} + \alpha H_{\mathrm{relax}}$, $\quad 0 \le \kappa \le .95,\ 0 \le \alpha \le 1.$

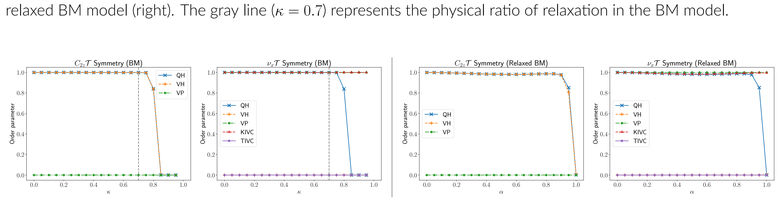


Numerical experiments are performed with 6×6 *discretization*, *spinless*, at *charge neutrality*. We use *average subtraction* to correct double counting. The initial states are the 5 exact ground states of the chiral model.

Results

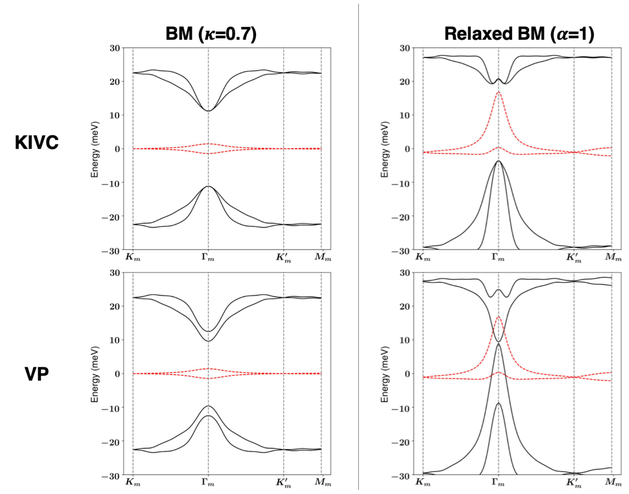


Energy difference to ground state (minimum of the five candidate states) per moiré site for the BM model (left) and relaxed BM model (right). The gray line ($\kappa = 0.7$) represents the physical ratio of relaxation in the BM model.



Symmetry order parameters $C_{2z}\mathcal{T}$ and $\nu_x\mathcal{T}$ for BM (left) and relaxed BM model (right).

Ground state HF band structure



Interacting band structure for ground state with physical parameters. BM model has insulating KIVC and VP states. Relaxed BM has a gap closing for VP state. (Black: HF band structure, Red: Non-interacting band structure)

Method: Interacting Model for TBG

Hartree-Fock Theory

A Slater determinant state $|\Psi_S\rangle$ with N_e electrons define a one-body reduced density matrix (1-RDM) by:

$$[P(\mathbf{k},\mathbf{k}')]_{mn} := \langle\Psi_S|\hat{f}^\dagger_{n\mathbf{k}'}\hat{f}_{m\mathbf{k}}|\Psi_S\rangle\,.$$

The Hartree-Fock ground state energy can be written as the minimization problem [6]

$$\mathcal{E}_{\mathrm{HF}} = \min \operatorname{tr}\Big((H_0 - H_{\mathrm{sub}} + J[P] - K[P])P\Big), \quad \text{s.t. } P^2 = P,\ \operatorname{tr}P = N_e. \tag{3}$$

$\hat{H}_0$ is the single particle dispersion (BM or relaxed BM).

The interacting terms consists of Coulomb (Hartree) and exchange (Fock) operator:

$$\hat{J}[P] = \frac{1}{|\Gamma_m^{\mathcal{K}}|}\sum_{\mathbf{k},\mathbf{k}'\in\mathcal{K}}\sum_{\mathbf{G}\in\mathcal{R}_{\mathrm{m}}^*}\sum_{m,n\in\mathcal{N}}\hat{V}(\mathbf{G})\operatorname{tr}\Big(\rho_{\mathbf{k}',\mathbf{k}'}(-\mathbf{G})P(\mathbf{k}')\Big)[\rho_{\mathbf{k},\mathbf{k}}(\mathbf{G})]_{mn}\hat{f}^\dagger_{m\mathbf{k}}\hat{f}_{n\mathbf{k}},$$

$$\hat{K}[P] = \frac{1}{|\Gamma_m^{\mathcal{K}}|}\sum_{\mathbf{k},\mathbf{k}'\in\mathcal{K}}\sum_{\mathbf{G}\in\mathcal{R}_{\mathrm{m}}^*}\sum_{m,n,m',n'\in\mathcal{N}}\hat{V}(\mathbf{k}'-\mathbf{k}+\mathbf{G})[\rho_{\mathbf{k},\mathbf{k}'}(\mathbf{G})]_{mn}[P(\mathbf{k}')]_{nm'}[\rho_{\mathbf{k}',\mathbf{k}}(-\mathbf{G})]_{n'm'}\hat{f}^\dagger_{m\mathbf{k}}\hat{f}_{n'\mathbf{k}}$$

Double gate screened potential $\hat{V}(\mathbf{q}') = \frac{2\pi}{\epsilon}\frac{\tanh(|\mathbf{q}'|d/2)}{|\mathbf{q}'|}$, with $\epsilon = 10.79$ and $d = 30$ nm.

Average subtraction to correct double counting e-e interaction $\hat{H}_{\mathrm{sub}} = \hat{J}[P_0] - \hat{K}[P_0]$, $P_0 \equiv \frac{1}{2}I_{4\times4}$.

Gauge Fixing and Initialization

Initialize calculations using five "ferromagnetic" Slater determinant states which are exact ground states at the chiral limit [7]. In the basis of $(\mathbf{K},\mathrm{A})$, $(\mathbf{K},\mathrm{B})$, $(\mathbf{K}',\mathrm{A})$, $(\mathbf{K}',\mathrm{B})$, the 1-RDM of these states are

$$P_{\mathrm{QH}} = \operatorname{diag}(1,0,0,1), \qquad P_{\mathrm{VH}} = \operatorname{diag}(1,0,1,0), \qquad P_{\mathrm{VP}} = \operatorname{diag}(1,1,0,0)$$

$$P_{\mathrm{KIVC}} = \frac{1}{2}\begin{bmatrix} 1 & & & -ie^{-i\varphi} \\ & 1 & ie^{-i\varphi} & \\ & -ie^{i\varphi} & 1 & \\ ie^{i\varphi} & & & 1 \end{bmatrix}, \qquad P_{\mathrm{TIVC}} = \frac{1}{2}\begin{bmatrix} 1 & & & e^{-i\varphi} \\ & 1 & e^{-i\varphi} & \\ & e^{i\varphi} & 1 & \\ e^{i\varphi} & & & 1 \end{bmatrix}.$$

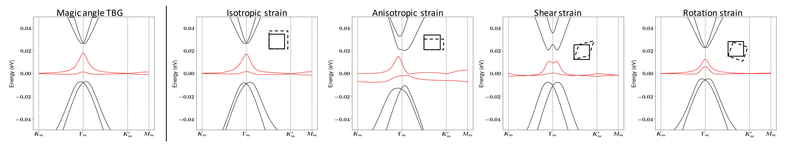
The symmetry order parameter of these candidate ground states are

	$O_{C_{2z}\mathcal{T}}$	$O_{\nu_x\mathcal{T}}$	$O_{\nu_y\mathcal{T}}$
QH	1	1	1
VH	1	0	0
VP	0	1	1

	$O_{C_{2z}\mathcal{T}}$	$O_{\nu_x\mathcal{T}}$	$O_{\nu_y\mathcal{T}}$
KIVC	$\lvert\operatorname{Re}(e^{i\varphi})\rvert$	1	0
TIVC	$\lvert\operatorname{Im}(e^{i\varphi})\rvert$	0	1

Conclusion

- Propose a novel many-body model of TBG which systematically accounts for the effects of structural relaxation.
- The relative ordering of candidate ground states is fairly consistent with simpler treatments of relaxation. Our model predicts the significantly earlier symmetry-breaking phase transitions.
- The framework of this work can also be adapted to include the effects of heterostrain



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