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2024

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University of California
Santa Barbara

Moiré Effects on Electrons and Phonons in Low Dimensional Materials

A dissertation submitted in partial satisfaction
of the requirements for the degree

Doctor of Philosophy
in
Materials

by

Wangqian Miao

Committee in charge:

Professor Susanne Stemmer, Co-Chair
Professor Xi Dai, Co-Chair
Professor Ram Seshadri
Professor Leon Balents

September 2024

The Dissertation of Wangqian Miao is approved.

Professor Ram Seshadri

Professor Leon Balents

Professor Xi Dai, Committee Chair

Professor Susanne Stemmer, Committee Chair

September 2024

Moiré Effects on Electrons and Phonons in Low Dimensional Materials

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Wangqian Miao

Dedicated to my parents and grandparents.

Acknowledgements

I still remember it was six years ago I finally made the decision to pursue a Ph.D. while on a train from Beijing to Nanjing. I feel very fortunate to have had Professor Xi Dai as my advisor for the past five years. His integrity and passion for physics have continuously inspired and encouraged me throughout this journey and will continue to do so in my future career. Over the years, we have explored several interesting topics across the field, which form the foundation of this dissertation. During this period, I spent around three years in Hong Kong and two years in Santa Barbara, enjoying the beautiful sunrises and sunsets by the sea while immersing myself in the study of physics. None of this would have been possible without his support and guidance.

I am also deeply grateful to have Professor Susanne Stemmer as my co-advisor at UCSB. She is an exceptional experimentalist in materials growth and transport measurement. I thoroughly enjoyed our discussions on the transport phenomena observed in the Cd_3As_2 system.

I extend my sincere thanks to Professor Ram Seshadri and Professor Leon Balents for serving on my Ph.D. advisory committee. Ram, thank you for arranging the wonderful office spaces at the Quantum Foundry in Elings Hall and for the memorable pizza night when Tianyu and I first arrived in Santa Barbara. Your kindness will always be remembered. Professor Leon Balents, your Physics 223 A course during my first quarter at UCSB is one of the best condensed matter physics courses I have ever taken.

The journey to a Ph.D. is never easy, especially during the pandemic. I want to thank all the group members, including Tianyu Qiao, Hao Shi, Shu Liang, Ren Zhang, Dr. Yuelin Shao, Dr. Song Sun, Dr. Shiyu Peng, and Dr. Hongbin Ren. Without their assistance, I could not have completed my Ph.D. studies. I also want to thank Dr. Chu

Li, Professor Jianpeng Liu, Professor Ding Pan, Professor Yao Wang, Professor Chaoxing Liu, and Professor Yulin Chen for their invaluable discussions on moiré phonons and electron-phonon couplings in the twisted bilayer graphene system. Special thanks to Dr. Binghao (Evan) Guo for his help during my time in Santa Barbara and for our ongoing discussions on the phenomena in Cd_3As_2 .

I am grateful to all my friends for their selfless help and encouragement. Especially during my time in Santa Barbara, it was always nice to go climbing, enjoy the amazing views of California, and visit Chinese restaurants on the weekends.

Lastly, I am thankful to have the best parents and grandparents in the world, who support all my decisions unconditionally. I love you all forever.

Curriculum Vitæ

Wangqian Miao

Education

- Sep 2021 – Sep 2024 Ph.D. in Materials (expected)
University of California, Santa Barbara, U.S.
Advisor: Prof. Susanne Stemmer & Prof. Xi Dai
- Sep 2019 – Jul 2021 M.Phil. in Physics
Hong Kong University of Science and Technology, Hong Kong
Advisor: Prof. Xi Dai
- Aug 2015 – Jul 2019 B.Sc. in Physics
Nanjing University, Nanjing, China

Publications

6. **Miao, W.**, Rashidi, A., Dai, X. (2024) "Artificial moiré engineering for an ideal BHZ model", arXiv:2409.08540.
5. Shi, H., **Miao, W.**, Dai, X., (2024) "Moiré optical phonons dancing with heavy electrons in magic-angle twisted bilayer graphene", arXiv:2402.11824.
4. Chen, C., Nuckolls, K.P., Ding, S., **Miao, W.**, Wong. D., Oh. M., Lee. R. L., He. S., Peng. C., Pei. D., Li. Y., Zhang. S., Liu. J., Liu. Z., Jozwiak. C., Bostwick. A., Rotenberg. E., Li. C., Han. X., Pan. D., Dai. X., Liu. C., Bernevig. B. A., Wang. Y., Yazdani. A., Chen. Y. (2023) "Strong Inter-valley Electron-Phonon Coupling in Magic-Angle Twisted Bilayer Graphene", arXiv:2303.14903.
3. **Miao, W.**, Guo, B., Stemmer, S., Dai, X. (2024) "Engineering the in-plane anomalous Hall effect in Cd_3As_2 thin films", *Phys. Rev. B.*, 109, 15, 155408.
2. Guo, B., **Miao, W.**, Huang, V., Lygo, A. C., Dai, X. Stemmer, S. (2023) "Zeeman Field-Induced Two-Dimensional Weyl Semimetal Phase in Cadmium Arsenide", *Phys. Rev. Lett.*, 131, 4, 046601.
1. **Miao, W.**, Li, C., Han, X., Pan, D., Dai, X. (2023) "Truncated atomic plane wave method for subband structure calculations of moiré systems", *Phys. Rev. B.*, 107, 12, 125112.

Abstract

Moiré Effects on Electrons and Phonons in Low Dimensional Materials

by

Wangqian Miao

Moiré materials have raised significant interest in recent years due to the observation of various intriguing phenomena, including novel superconductivity, the quantum anomalous Hall effect, and its fractional version. However, there are two significant challenges in the study of moiré materials. The first challenge is to accurately and efficiently present the low-energy band structure in such large systems. The second challenge is to find new materials where inhomogeneity caused by strain and twist has minimal impact on experiments.

In the first part of this dissertation, I discuss a newly developed numerical method called the Truncated Atomic Plane Wave (TAPW) method, which addresses the first challenge. For twisted bilayer graphene, this method serves as a natural extension of the original Bistritzer and MacDonald model, incorporating corrections for higher-order Fourier components and non-local tunneling, by projecting the full tight binding Hamiltonian on a series of plane waves. Additionally, we combine this method with numerical atomic orbital-based density functional theory to present the valley-resolved band structure of moiré transition metal dichalcogenide (TMD) materials. By drawing an analogy between the tight-binding formalism of electrons and phonons, we solve the problem of the low-frequency moiré phonons and systematically study the effective coupling between flat band electrons and transverse optical phonons in the magic-angle twisted bilayer graphene (MATBG) system.

The second part of this dissertation focuses on the second challenge, primarily proposing a new platform to engineer moiré physics. Here, I focus on another representation of low-dimensional materials: topological thin films, with a special emphasis on high quality Dirac semimetal Cd_3As_2 . The magneto transport properties of Cd_3As_2 thin films were meticulously studied, revealing a possible Weyl semimetal phase and a quantum anomalous Hall phase both induced by in-plane Zeeman coupling. The (001) grown Cd_3As_2 thin film at critical thickness serves as an experimental realization of the Bernevig-Hughes-Zhang (BHZ) model. In light of recent experimental progress in superlattice and gate engineering, the emergent band topology in the BHZ model with superlattice potential is discussed, and an interaction driven Chern insulator phase can be available by spontaneously breaking time reversal symmetry.

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Chapter 1

Introduction

1.1 Moiré materials, a new playground

With the continuous advancement of technology and scientific understanding, researchers have made significant progress in the field of low-dimensional materials. These materials exhibit intriguing properties due to quantum confinement, leading to a wide range of experimental findings. Some notable discoveries include the quantum Hall effect [5], fractional quantum Hall effect [6], and their anomalous versions [7] even in the absence of an external magnetic field.

Low-dimensional materials can generally be classified into two main groups based on their properties and synthesis methods. The first group consists of a large family of materials that can be grown on suitable substrates using techniques such as molecular beam epitaxy (MBE). These materials typically have thicknesses on the order of tens of nanometers, and they allow the creation of clean two-dimensional electron gases. These systems have been extensively studied, and researchers have been able to explore their unique electronic and transport properties, such as high mobility, ballistic trans-

port, and quantum confinement effects. The second group comprises two-dimensional van der Waals (vdW) materials. These materials are composed of individual atomic layers that are weakly bonded together by van der Waals forces. Graphene [8], a single layer of carbon atoms arranged in a honeycomb lattice, is one of the most well-known examples in this category. Other van der Waals materials, such as transition metal dichalcogenides (TMDs) and black phosphorus, have also gained significant attention. These materials exhibit diverse physical properties, including tunable bandgaps, strong light-matter interactions, and novel spin and valley physics, making them promising candidates for various applications in electronics and quantum technologies.

Two-dimensional van der Waals (vdW) materials provide an effective approach to engineer their physical properties by creating complex heterostructures. These heterostructures introduce additional degrees of freedom, including stacking configurations, twist angles, and lattice mismatches. These variations result in a new periodicity known as a moiré scale periodicity. The moiré superlattice leads to band folding in momentum space, and the interlayer interaction and lattice relaxation further complicate the electronic structure.

In 2010, it was first predicted that twisted bilayer graphene [9] near the magic angle could exhibit flat bands, where electron-electron interactions are significantly enhanced. This prediction was later confirmed in 2018 [10, 11] and this discovery sparked extensive research in moiré materials. Recent experimental advancements have unveiled novel superconductivity [11, 12], Mott insulator behavior [10], quantum anomalous Hall effect [13, 14] and even its fractional version [1, 15, 16]. These intriguing phenomena has strong connections with Berry curvature. Berry curvature plays a central role in various phenomena observed in quantum materials, including the anomalous Hall effect, valley Hall effect and the concept of topological invariants.

This geometrical quantity describes the topological properties of the electron wavefunctions of the material. Additionally, other band-geometric quantities such as the quantum metric, orbital magnetic moment, and Berry curvature dipole contribute to interesting Hall responses in transport and optics measurements.

Undoubtedly, two-dimensional moiré materials have opened up new opportunities for exploring Berry physics by providing various tuning knobs such as twist angles, lattice configurations, and external electrical fields. This unique class of materials combines momentum-space topology with strong correlation physics, offering a versatile platform for investigating fundamental phenomena. In the subsequent section of this introduction, we will delve into the fundamental concepts and numerical methods used to study band topology. Additionally, we will present two illustrative examples: a ferromagnetic Kagome lattice and twisted MoTe_2 . In both systems, band topology is of great importance in understanding their intriguing properties and behaviors.

1.2 Basic concepts and techniques

1.2.1 Tight binding formalism

In order to comprehend the physical properties of materials and their potential response to external fields, it is necessary to construct a low-energy Hamiltonian under certain approximations. The tight binding method is a powerful tool that assists physicists in establishing connections between general theories and specific scenarios.

We begin by formulating the tight-binding description in solid-state systems. Due to the presence of translational symmetry, it is possible to construct a basis known as

the tight-binding basis using a Bloch sum:

$$\psi_{an}(\mathbf{k}) = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot (\mathbf{R} + \boldsymbol{\tau}_a)} \phi_n(\mathbf{r} - \mathbf{R} - \boldsymbol{\tau}_a), \quad (1.1)$$

where a denotes the atom coordinate and n represents the n -th localized orbital. Taking advantage of the Bloch theorem, the matrix element of the tight binding Hamiltonian is

$$\begin{aligned} H_{anbm}(\mathbf{k}) &= \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot (\mathbf{R} + \boldsymbol{\tau}_b - \boldsymbol{\tau}_a)} \langle \phi_n(\mathbf{r} - \mathbf{0} - \boldsymbol{\tau}_a) | H | \phi_m(\mathbf{r} - \mathbf{R} - \boldsymbol{\tau}_b) \rangle \\ &= \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot (\mathbf{R} + \boldsymbol{\tau}_b - \boldsymbol{\tau}_a)} E_{anbm}(\boldsymbol{\tau}_a, \mathbf{R} + \boldsymbol{\tau}_b), \end{aligned} \quad (1.2)$$

where $E_{anbm}(\boldsymbol{\tau}_a, \mathbf{R} + \boldsymbol{\tau}_b)$ represents the hopping matrix element between the n -th orbital located at site a and the m -th orbital located at site b . The band structure of the system and the corresponding wavefunctions can be obtained by diagonalizing the Hamiltonian.

In the early days of calculating the band structures of typical semiconductors, it was observed that preserving atomic orbitals on each site could reasonably reproduce the band structure. Empirical determination of hopping parameters, often using a Slater-Koster (SK) formalism [17], was employed to describe the hopping between neighboring sites. Additionally, the spin-orbit coupling (SOC) effect emerged as a significant factor influencing the behavior of various materials. In the tight-binding formalism, when (pseudo) atomic orbitals are chosen as the basis functions, the SOC effect can be directly incorporated through ad hoc calculations. The onsite SOC term, denoted as $H_{\text{SOC}} = \lambda \mathbf{L} \cdot \mathbf{S}$, is directly added to the original tight-binding Hamiltonian, with λ representing the strength of the SOC, which can be fitted using first-principles calculations. This inclusion of the SOC term allows the tight-binding model to account for the coupling between the orbital angular momentum ($\mathbf{L}$) and the spin angular momentum

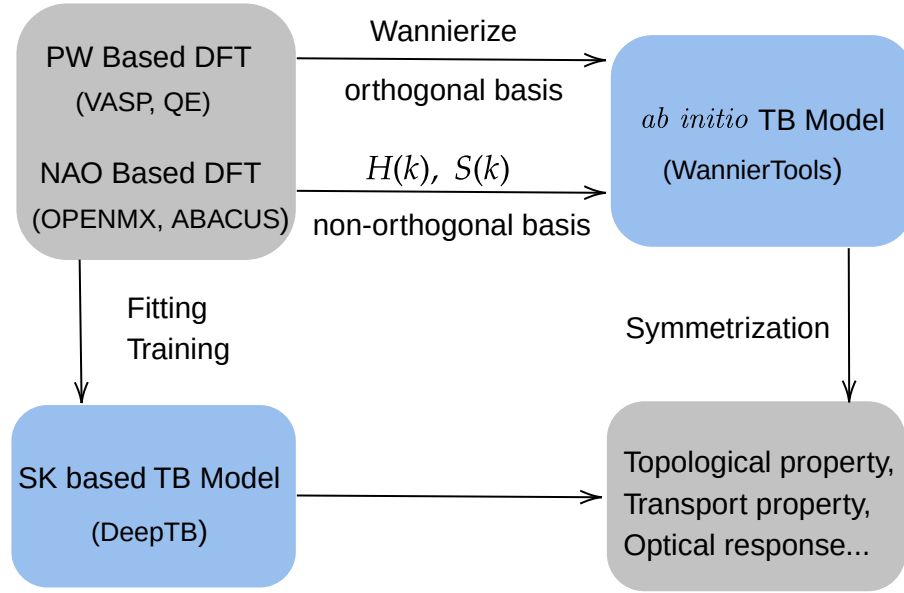


Figure 1.1: Road map for the tight binding Models in modern condensed matter physics. Corresponding computational software is also listed. “PW based DFT” represents plane wave based density functional theory and “NAO based DFT” represents numerical atomic orbital based density functional theory.

(S) of the electrons, providing a more accurate description of the electronic properties of materials with strong SOC. However, recent advancements in deep neural networks (DNNs) and first-principles calculations have significantly expanded the capabilities of the tight binding method.

Two notable developments have revolutionized the field of tight-binding calculations, see the road map for tight binding models in Fig. 1.1. The first development involves the parameterization of SK tight-binding models. Traditionally, these parameters were determined empirically or through fitting to experimental data. However, with the advent of DNNs, it has become possible to *ab initio*ly determine SK parameters by training them on datasets generated from first-principles calculations, as demonstrated by methods like DeepTB [18].

The second development relates to the extension of the basis functions used in tight-

binding calculations. The wavefunctions, denoted by ϕ , can be represented as either Wannierized numerical orbitals [19, 20] or pseudo atomic orbitals [21]. Wannierized numerical orbitals have become widely employed in modern computational physics and are integrated into software packages such as Wannier90 [22]. On the other hand, pseudo atomic orbitals can be directly combined with density functional theory (DFT) and are implemented in several DFT packages such as OPENMX [21] and ABACUS [23]. It is worth noting that numerical atomic orbitals do not necessarily need to be orthogonalized. Consequently, the introduction of an overlap matrix becomes necessary to account for the non-orthogonality of these orbitals,

$$\begin{aligned} S_{anbm}(\mathbf{k}) &= \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot (\mathbf{R} + \boldsymbol{\tau}_b - \boldsymbol{\tau}_a)} \langle \phi_n(\mathbf{r} - \mathbf{0} - \boldsymbol{\tau}_a) | \phi_m(\mathbf{r} - \mathbf{R} - \boldsymbol{\tau}_b) \rangle \\ &= \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot (\mathbf{R} + \boldsymbol{\tau}_b - \boldsymbol{\tau}_a)} O_{anbm}(\boldsymbol{\tau}_a, \mathbf{R} + \boldsymbol{\tau}_b), \end{aligned} \quad (1.3)$$

Here, $O_{anbm}(\boldsymbol{\tau}_a, \mathbf{R} + \boldsymbol{\tau}_b)$ represents the overlap between two orbitals. The eigenstates and eigenvalues of the system can be obtained by solving a generalized eigenvalue problem. When we obtain the TB models for a material, we can use these models to study its physical properties, including topological property, transport behaviour and optical response. By investigating these properties, we gain insights into the behavior of the material and its potential applications. These studies also guide experimental research and contribute to the development of new devices and technologies.

We then investigate the symmetry constraints for the tight binding Hamiltonian [24] and corresponding hopping matrix. For a general unitary symmetry operation g (does not contain time reversal $\mathcal{T}$), we have the following statement,

$$g\phi_n(\mathbf{r}) = \phi_n(g^{-1}\mathbf{r}) = \phi_m(\mathbf{r})D_{mn}(g). \quad (1.4)$$

where $D(g)$ is the representation matrix under basis ϕ . In quantum mechanics, we say symmetry which indicates this operation commutes with the Hamiltonian $gH = Hg \rightarrow g^{-1}Hg = H$. For the matrix element of the hopping matrix, one can check that,

$$\begin{aligned} E_{anbm}(\tau_a, \mathbf{R} + \tau_b) &= \langle \phi_n(\mathbf{r} - \mathbf{0} - \tau_a) | g^{-1} H g | \phi_m(\mathbf{r} - \mathbf{R} - \tau_b) \rangle \\ &= D_{nk}(g^{-1}) \langle \phi_k(\mathbf{r} - g(\mathbf{0} + \tau_a)) | H | \phi_\ell(\mathbf{r} - g(\mathbf{R} + \tau_b)) \rangle D_{\ell m}(g) \quad (1.5) \\ &= D_{nk}(g^{-1}) E_{akbl}(g\tau_a, g(\mathbf{R} + \tau_b)) D_{\ell m}(g). \end{aligned}$$

In the matrix form, it is $E(\tau_a, \mathbf{R} + \tau_b) = D(g^{-1}) E(g\tau_a, g(\mathbf{R} + \tau_b)) D(g)$. This symmetry constraint is much easier to check when using the realistic tight binding model and it is also implemented in WannierTools [25]. For the tight binding Hamiltonian, we see,

$$\begin{aligned} H_{anbm}(\mathbf{k}) &= \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot (\mathbf{R} + \tau_b - \tau_a)} \langle \phi_n(\mathbf{r} - \mathbf{0} - \tau_a) | g^{-1} H g | \phi_m(\mathbf{r} - \mathbf{R} - \tau_b) \rangle \\ &= D_{nk}(g^{-1}) H_{akbl}(g\mathbf{k}) D_{\ell m}(g). \end{aligned} \quad (1.6)$$

It is important to note that when g is unitary, $D(g^{-1}) = D^\dagger(g)$. In matrix form, the equation can be written as $H(\mathbf{k}) = D(g^{-1}) H(g\mathbf{k}) D(g)$. This result is particularly useful when constructing the symmetry-adapted $k \cdot p$ Hamiltonian [26].

When an operation g contains the time reversal operator $\mathcal{T}$, it can be expressed as $g = U\mathcal{K}$, where U represents the unitary part of the operation, and $\mathcal{K}$ denotes the complex conjugation operation. In this case, we can verify the following statement,

$$\begin{aligned} E(\tau_a, \mathbf{R} + \tau_b) &= D(U^{-1}) E^*(U\tau_a, U(\mathbf{R} + \tau_b)) D(U) \\ H(\mathbf{k}) &= D(U^{-1}) H^*(-U\mathbf{k}) D(U). \end{aligned} \quad (1.7)$$

We can use a minimal example to help understand the aforementioned analysis. Consider a square lattice with an s orbital on each site. When the system is at half-

filling, the corresponding tight binding Hamiltonian can be expressed as follows:

$$H = \sum_{\langle ij \rangle} t_{ij} c_i^\dagger c_j + J \sum_i \mathbf{S}_i \sigma^{\alpha\beta} c_{i\alpha}^\dagger c_{i\beta}. \quad (1.8)$$

The second term in the above Hamiltonian arises from the exchange interaction J and can be interpreted as a decoupled Heisenberg model with hopping terms t_{ij} under the mean-field approximation. Here, α and β are spin indices, and $\mathbf{S}_i$ represents the spin configuration at each site. The basis function in this case is $s \otimes (\uparrow, \downarrow)$.

The C_{2z} rotational operator can be expressed as $D(C_{2z}) = e^{-i\pi/2\sigma_z}$, and the time reversal operator is set as $D(\mathcal{T}) = -i\sigma_y\mathcal{K}$. Assuming a ferromagnetic system, when all spins are aligned out of the plane, we can observe that the $C_{2z}\mathcal{T}$ symmetry is broken. However, when all spins lie in the plane, the $C_{2z}\mathcal{T}$ symmetry survives. For the $C_{2z}\mathcal{T}$ symmetry, the unitary part of the representation matrix can be chosen as $D(C_{2z}\mathcal{T}) = -ie^{-i\pi/2\sigma_z}\sigma_y$. The above analysis can be quickly confirmed using the following calculations,

$$\begin{aligned} D(C_{2z}\mathcal{T})^{-1}\sigma_x^*D(C_{2z}\mathcal{T}) &= \sigma_x \\ D(C_{2z}\mathcal{T})^{-1}\sigma_y^*D(C_{2z}\mathcal{T}) &= \sigma_y \\ D(C_{2z}\mathcal{T})^{-1}\sigma_z^*D(C_{2z}\mathcal{T}) &= -\sigma_z. \end{aligned} \quad (1.9)$$

1.2.2 Berry curvature and band topology

Berry curvature is a fundamental concept in condensed matter physics that plays a crucial role in understanding the behavior of electrons in materials. It arises from the geometric phase acquired by an electron wavefunction as it evolves along a closed path in the momentum space. The Berry curvature is associated with the geometry of the energy bands and can give rise to intriguing phenomena such as the quantum Hall effect

and topological insulators. It provides valuable insights into the electronic structure and transport properties of materials, offering a powerful tool for studying and predicting novel quantum phenomena. In this context, investigating the Berry curvature has become an active area of research, with applications spanning from fundamental condensed matter physics to the design of novel electronic devices. The definition of the quantum geometric tensor (for wave function ψ_λ) is:

$$Q_{\mu\nu} = \langle \partial_\mu \psi_\lambda | \partial_\nu \psi_\lambda \rangle - \langle \partial_\mu \psi_\lambda | \psi_\lambda \rangle \langle \psi_\lambda | \partial_\nu \psi_\lambda \rangle \quad (1.10)$$

Note that,

$$\langle \psi_m | \partial_\mu (H | \psi_n \rangle) = \langle \psi_m | \partial_\mu H | \psi_n \rangle + \langle \psi_m | H | \partial_\mu \psi_n \rangle \quad (1.11)$$

Given that the LHS equals to $E_n \langle \psi_m | \partial_\mu \psi_n \rangle$ and the RHS equals to $\langle \psi_m | \partial_\mu H | \psi_n \rangle + E_m \langle \psi_m | \partial_\mu \psi_n \rangle$, we arrive:

$$\langle \psi_m | \partial_\mu \psi_n \rangle = \frac{\langle \psi_m | \partial_\mu H | \psi_n \rangle}{E_n - E_m} \quad (1.12)$$

Then the quantum metric tensor should be:

$$Q_{\mu\nu} = \langle \partial_\mu \psi_\lambda | (1 - |\psi_\lambda\rangle \langle \psi_\lambda|) | \partial_\nu \psi_\lambda \rangle \quad (1.13)$$

For band n , the above equation can be further simplified:

$$Q_{\mu\nu} = \langle \partial_\mu \psi_n | \sum_{m \neq n} (|\psi_m\rangle \langle \psi_m|) | \partial_\nu \psi_n \rangle \quad (1.14)$$

When the system is non-degenerate, for the solid state system, the quantum geometric tensor for band n is defined as,

$$\mathcal{Q}_{ij}(\mathbf{k}) = \sum_{m \neq n} \frac{v_{nm}^i(\mathbf{k}) v_{mn}^j(\mathbf{k})}{(\varepsilon_n(\mathbf{k}) - \varepsilon_m(\mathbf{k}))^2}, \quad (1.15)$$

where $v_{nm}^i(\mathbf{k}) = \langle u_{n\mathbf{k}} | \partial_i H_{\mathbf{k}} | u_{m\mathbf{k}} \rangle$ represents fore matrix element of the velocity operator and ij are coordinate indices. The so called quantum metric tensor $\mathcal{G}_{ij} = \text{Re } \mathcal{Q}_{ij}$ and Berry curvature tensor $\Omega_{ij} = -2\text{Im } \mathcal{Q}_{ij}$. More explicitly,

$$\Omega_{ij}(\mathbf{k}) = i \sum_{m \neq n} \frac{v_{nm}^i(\mathbf{k}) v_{mn}^j(\mathbf{k}) - v_{mn}^i(\mathbf{k}) v_{nm}^j(\mathbf{k})}{(\varepsilon_n(\mathbf{k}) - \varepsilon_m(\mathbf{k}))^2}. \quad (1.16)$$

The above expression for the Berry curvature is consistent with the TKNN formula derived from linear response theory. The velocity operator is hermitian ($\mathbf{v}(\mathbf{k}) = \nabla_{\mathbf{k}} H_{\mathbf{k}} = [\mathbf{v}(\mathbf{k})]^\dagger$) and the second part in the numerator is the complex conjugate of the first part.

Now, let's discuss the symmetry constraints for the Berry curvature. We will examine the constraints imposed by time reversal symmetry ($\mathcal{T}$), space inversion symmetry ($\mathcal{P}$), space-time inversion symmetry ($C_{2z}\mathcal{T}$), and general space rotation ($\mathcal{R}$) with a focus on rotation around the z axis. For the Hamiltonian, we have the following statement,

$$\begin{aligned} \mathcal{P} : H(\mathbf{k}) &= D^\dagger H(-\mathbf{k}) D, \\ \mathcal{T} : H(\mathbf{k}) &= D^\dagger H^*(-\mathbf{k}) D, \\ \mathcal{R} : H(\mathbf{k}) &= D^\dagger H(g^{-1}\mathbf{k}) D, \\ C_{2z}\mathcal{T} : H(k_x, k_y, k_z) &= D^\dagger H^*(k_x, k_y, -k_z) D, \\ \mathcal{PT} : H(\mathbf{k}) &= D^\dagger H^*(\mathbf{k}) D, \end{aligned} \quad (1.17)$$

where D represents the unitary part of the representation matrix. We can further

derive constraints for the energy dispersions and wavefunctions. Based on the above discussion, we can readily confirm the following equations,

$$\begin{aligned}
\mathcal{P} : \varepsilon_{n\mathbf{k}} &= \varepsilon_{n-\mathbf{k}}, \quad |u_{n\mathbf{k}}\rangle \sim |u_{n-\mathbf{k}}\rangle \\
\mathcal{T} : \varepsilon_{n\mathbf{k}} &= \varepsilon_{n-\mathbf{k}}, \quad |u_{n\mathbf{k}}\rangle \sim |u_{n-\mathbf{k}}^*\rangle \\
C_{2z}\mathcal{T} : \varepsilon_{n\mathbf{k},q_z} &= \varepsilon_{n\mathbf{k},-q_z}, \quad |u_{n\mathbf{k},q_z}\rangle \sim |u_{n\mathbf{k},-q_z}^*\rangle,
\end{aligned} \tag{1.18}$$

and $\sim$ represents two wave functions can be connected through a unitary transformation. For the velocity operator and Berry curvature we can check that,

$$\begin{aligned}
\mathcal{P} : \langle u_{n\mathbf{k}} | \partial_i H_{\mathbf{k}} | u_{m\mathbf{k}} \rangle &= -\langle u_{n-\mathbf{k}} | \partial_{-\mathbf{k}_i} H_{-\mathbf{k}} | u_{m-\mathbf{k}} \rangle = -v_{nm}^i(-\mathbf{k}) \\
&\rightarrow \Omega_{ij}(\mathbf{k}) = \Omega_{ij}(-\mathbf{k}) \\
\mathcal{T} : \langle u_{n\mathbf{k}} | \partial_i H_{\mathbf{k}} | u_{m\mathbf{k}} \rangle &= -(\langle u_{n-\mathbf{k}} | \partial_{-\mathbf{k}_i} H_{-\mathbf{k}} | u_{m-\mathbf{k}} \rangle)^* = -v_{mn}^i(-\mathbf{k}) \\
&\rightarrow \Omega_{ij}(\mathbf{k}) = -\Omega_{ij}(-\mathbf{k}) \\
C_{2z}\mathcal{T} : \langle u_{n\mathbf{k},k_z} | \partial_i H_{\mathbf{k}} | u_{m\mathbf{k},-k_z} \rangle &= (\langle u_{n\mathbf{k},-k_z} | \partial_i H_{\mathbf{k},-k_z} | u_{m\mathbf{k},-k_z} \rangle)^* = v_{mn}^i(\mathbf{k}, -k_z) \\
&\rightarrow \Omega_{ij}(k_x, k_y, k_z) = -\Omega_{ij}(k_x, k_y, -k_z)
\end{aligned} \tag{1.19}$$

The constraints on the Berry curvature arising from rotational symmetry can be complicated. For a general rotation (or rotation+reversion) g of a vector $\mathbf{k}$: $(g\mathbf{k})_i = \sum_j g_{ij}k_j$, the following transformation rule is useful,

$$\partial_{\mathbf{k}} = g^T \partial_{g\mathbf{k}}, \tag{1.20}$$

which results from the chain rule, $\partial_{\mathbf{k}_j} = \sum_i \frac{\partial (g\mathbf{k})_i}{\partial k_j} \partial_{(g\mathbf{k})_i} = \sum_i g_{ij} \partial_{(g\mathbf{k})_i}$. Then constraints results from the rotational symmetry is,

$$g(C_{nz}) : H(\mathbf{k}) = U H(g^{-1}\mathbf{k}) U^\dagger \rightarrow \varepsilon_{n\mathbf{k}} = \varepsilon_{ng^{-1}\mathbf{k}}, |u_{n\mathbf{k}}\rangle \sim |u_{ng^{-1}\mathbf{k}}\rangle. \tag{1.21}$$

Specifically, for the velocity operator:

$$\begin{aligned}
\mathbf{v}_{nm}(\mathbf{k}) &= \langle u_{n\mathbf{k}} | \partial_{\mathbf{k}} H(\mathbf{k}) | u_{m\mathbf{k}} \rangle \\
&= \langle u_{ng^{-1}\mathbf{k}} | \partial_{\mathbf{k}} H(g^{-1}\mathbf{k}) | u_{mg^{-1}\mathbf{k}} \rangle \\
&= \langle u_{ng^{-1}\mathbf{k}} | (g^{-1})^T \partial_{g^{-1}\mathbf{k}} H(g^{-1}\mathbf{k}) | u_{mg^{-1}\mathbf{k}} \rangle \\
&= (g^{-1})^T \mathbf{v}_{nm}(g^{-1}\mathbf{k}).
\end{aligned} \tag{1.22}$$

Indeed, considering the constraints mentioned earlier, it is obvious that the Berry curvature Ω_{xy} will exhibit rotational invariance.

We now start to consider the symmetry effects on the Berry curvature by using the example of $C_{2z}\mathcal{T}$ symmetry in two dimensions. When there is no band degeneracy, this local symmetry results in a trivial Berry curvature. By examining the minimal magnetic space group (MSG) 3.3 (BNS symbol), which includes identity (E) and $C_{2z}\mathcal{T}$ operations, we find that the representation matrix for $C_{2z}\mathcal{T}$ can be chosen as $\text{diag}(1, 1)\mathcal{K}$. Under this constraint, a tilted massive Dirac fermion serves as the minimal model Hamiltonian,

$$H = v(k_x + k_y)\sigma_x + vk_z\sigma_y + (m + tk_x)\sigma_z, \tag{1.23}$$

where v is the Fermi velocity, m will gap out the Dirac points and t is responsible for tilting the dispersion. The Berry curvature for a two level system $H = \mathbf{h} \cdot \boldsymbol{\sigma}$ is:

$$\Omega_{ij} = \frac{1}{2|\mathbf{h}|^3} \epsilon_{abc} h_c \partial_i h_a \partial_j h_b. \tag{1.24}$$

For this minimal model, we can write down the expression for Berry curvature,

$$\Omega_{xy} = \frac{tv^2(m + tk_x)^2 k_z}{((m + tk_x)^2 + v^2(k_x + k_y)^2 + v^2 k_z^2)^{\frac{5}{2}}}. \quad (1.25)$$

We can check $\Omega_{xy}(k_z) = -\Omega_{xy}(-k_z)$. Given that $\varepsilon(k_z) = \varepsilon(-k_z)$, the Berry curvature's contribution to the anomalous Hall effect σ_{xy} will cancel out.

1.3 Anomalous Hall effect

The quantum Hall (QH) effect is characterized by the quantization of the Hall conductance in materials at integer multiples of e^2/h . This effect, observed in two-dimensional electron gas systems subjected to strong magnetic fields, has been crucial in metrology due to its high precision. The phenomenon of precise quantization of Hall resistance raised the question of whether it could occur without a magnetic field. In 1982, the TKNN formula [27] established a connection between the Hall conductivity σ_{xy} for a band insulator and the Berry curvature $\Omega(\mathbf{k})$ in momentum space. This quantized value is the Chern number,

$$\mathcal{C} = -\frac{1}{2\pi} \sum_{n \in \text{occ.}} \int_{\text{B.Z.}} \Omega_n(\mathbf{k}) d\mathbf{k}, \quad (1.26)$$

where n is a band index and the Hall conductance is ,

$$\sigma_{xy} = \frac{e^2}{h} \mathcal{C}. \quad (1.27)$$

This formula revealed that the Hall conductance is quantized as an integer multiple of 2π and is experimentally observable. While the TKNN formula applies to electrons in

periodic potentials, the concept of non-zero Chern numbers in the absence of a magnetic field was emphasized in a significant paper by Haldane [28]. The terms “Chern insulator” and “quantum anomalous Hall (QAH) insulator” are used interchangeably to describe band insulators carrying non-zero Chern number. The anomalous Hall (AH) effect in magnetic conductors and the role of momentum-space Berry phases in transport are explained by the modern Berry phase theory [29]. The TKNN formula predicts quantized Hall conductance in 2D magnetic insulators and semiconductors, but the quantized value is almost always zero without a magnetic field.

The realization of the QAH effect was initially challenging until the discovery of topological insulators (TIs) in 2006 [30, 31]. Adding magnetism to TIs through the introduction of magnetic dopants led to the successful realization of the QAH effect [32]. Recent progress has also been made in achieving the QAH effect in materials with intrinsic magnetism and moiré superlattice. These materials are summarized as follows,

- Magnetic doped TI:

Cr doped $(\text{Bi}_2, \text{Sb}_2)\text{Te}_3$ [32], V doped $(\text{Bi}_2, \text{Sb}_2)\text{Te}_3$ [33].

- Intrinsic magnetic insulators:

MnBi_2Te_4 flakes [34].

- Moiré superlattice:

Twisted bilayer graphene [13], $\text{MoTe}_2/\text{WSe}_2$ [14], twisted MoTe_2 [1, 16], rhombohedral graphene systems [35–37].

In the subsequent section, we will explore two examples where the Quantum Anomalous Hall (QAH) effect can be observed, but through different mechanisms.

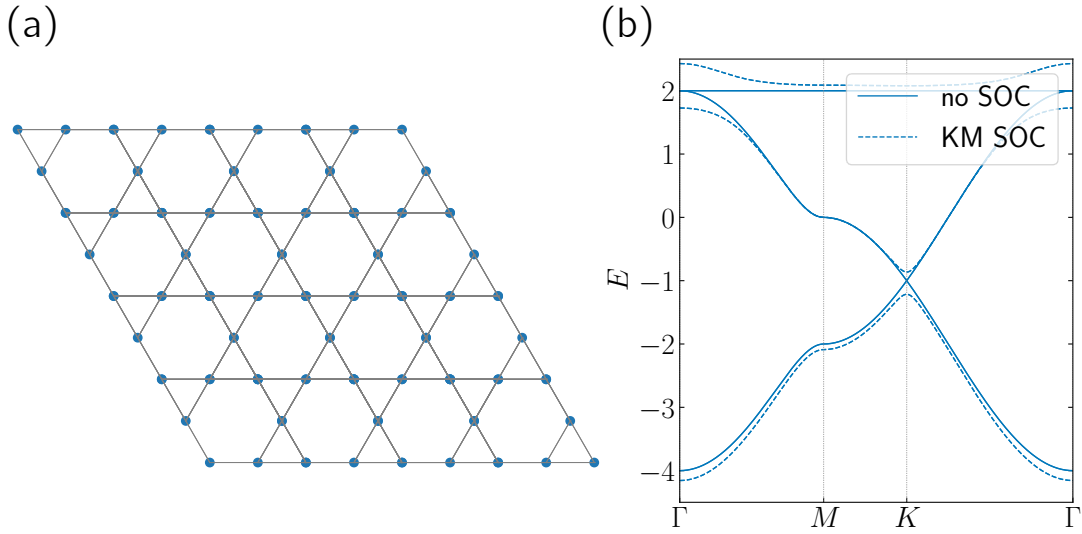


Figure 1.2: (a) Lattice geometry of a Kagome lattice. (b) Energy bands of the Kagome lattice with nearest neighbor hopping conserved ($t_{ij} = 1$). The introduction of Kane-Mele (KM) type spin-orbit coupling (SOC) $\lambda_1 = 0.3$ leads to the gapping out of the Dirac points and all bands are doubly degenerate due to Kramers degeneracy.

1.3.1 Kagome ferromagnets

In this section, we will provide a brief overview of a tight-binding model for the Kagome lattice, which can shed further light on the previously discussed concepts. The Kagome lattice has attracted significant attention in recent years due to the discovery of unconventional superconductivity in certain Kagome-based systems [38, 39]. This has spurred extensive research focused on understanding the unique properties exhibited by Kagome systems, including investigations into superconductivity [40, 41], charge density wave orders [42, 43], and the giant anomalous Hall effect [44].

The Kagome lattice is a two-dimensional lattice structure characterized by a repeating pattern of corner-sharing triangles, as shown in Fig. 1.2-(a). It consists of interconnected vertices forming hexagons, with each hexagon enclosing a central triangular unit. The lattice exhibits a hexagonal symmetry and possesses a honeycomb sublattice

structure. In the Kagome lattice, each lattice site is surrounded by three nearest neighboring sites, forming a highly interconnected network. This unique geometry gives rise to interesting electronic properties, such as geometric frustration and topological effects [45]. The tight binding Hamiltonian for a Kagome lattice with s orbitals on site ($s \otimes (\uparrow, \downarrow)$) can be written down,

$$H = \sum_{\langle ij \rangle} t_{ij} c_i^\dagger c_j + J \sum_i \mathbf{S}_i \sigma^{\alpha\beta} c_{i\alpha}^\dagger c_{i\beta} + i\lambda_1 \sum_{\langle ij \rangle} v_{ij} \sigma_z c_i^\dagger c_j + i\lambda_2 \sum_{\langle ij \rangle} [\mathbf{e}_{ij} \times \boldsymbol{\sigma}]_z c_i^\dagger c_j \quad (1.28)$$

The Hamiltonian consists of several terms that describe different physical interactions in the system. The first term accounts for electron hopping (t_{ij}) between neighboring sites, representing their movement and kinetic energy. The second term represents the mean-field exchange interaction (J), describing how electrons interact with each other. This term provides a simplified description of the collective behavior of electrons.

The third term in the Hamiltonian corresponds to Kane-Mele spin-orbit coupling (SOC) where $v_{ij} = -v_{ji} = 1$ [46]. It preserves the electron spin in the z direction, and it is associated with the quantum spin Hall effect (QSH) [30, 46]. The QSH effect leads to the emergence of topologically protected edge states in two-dimensional materials with time-reversal symmetry. Kane-Mele SOC typically arises from intrinsic crystal symmetry, which is why it is considered only in the second nearest neighbor interactions in the case graphene (inversion symmetry forbids the nearest neighbour KM-SOC).

The fourth term accounts for Rashba spin-orbit coupling [47] and $\mathbf{e}_{ij}$ denotes the vector connecting two atoms. This SOC couples the electron spin to its in-plane momentum and arises from the breaking of inversion symmetry, often induced by an exter-

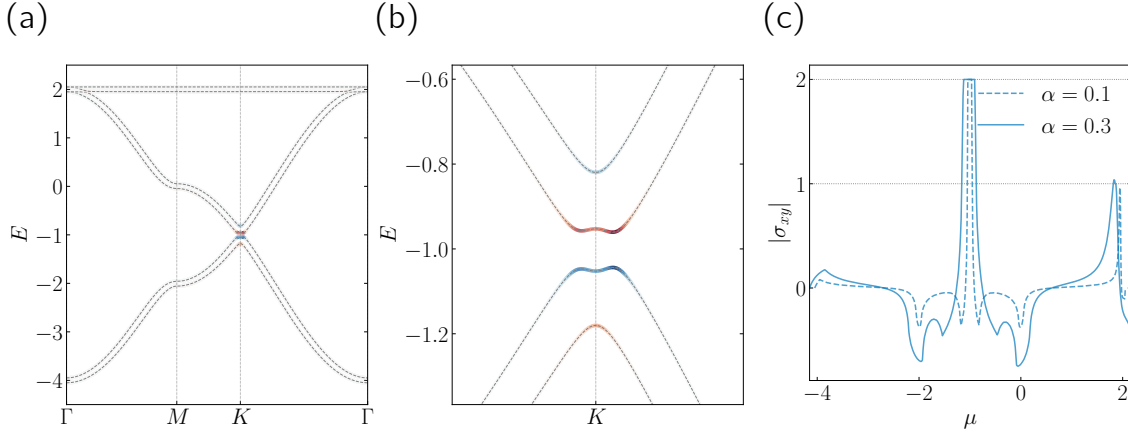


Figure 1.3: (a) Energy bands of a ferromagnetic Kagome lattice with Rashba-type spin-orbit coupling (SOC). The Rashba SOC opens a gap near the Dirac points, and the presence of magnetism breaks time reversal symmetry. These effects result in the formation of a Chern insulator. (b) Zoomed-in energy bands with depicted Berry curvature in (a). Different colors indicate the sign of the Berry curvature. The Berry curvature is concentrated around the original Dirac points. (c) Calculated anomalous Hall transport curve showing distinct plateaus ($\alpha = \lambda_2/t_{ij} = J/t_{ij}, t_{ij} = 1$). Two plateaus indicate the presence of Chern insulators with Chern numbers $\mathcal{C} = -2$ and $\mathcal{C} = -1$, obtained by tuning the chemical potential.

nal electric field¹. It is important to note that both types of SOC respect time-reversal symmetry, as confirmed by the equation $\mathcal{T}^{-1}i\sigma\mathcal{T} = i\sigma$.

Quantum anomalous Hall effect

In the absence of spin-orbit coupling (SOC) and exchange interaction in the Hamiltonian, the Kagome lattice exhibits flat bands and Dirac cones in its energy spectrum which is depicted in Fig. 1.2-(b). These features arise due to the lattice geometry and result in interesting electronic properties. Additionally, the bare Hamiltonian possesses $\mathcal{PT}$ symmetry, leading to doubly degenerate energy bands known as Kramers degeneracy.

¹In this case, this statement can be quickly checked through $E(r_i, r_j) \neq E(-r_i, -r_j)$.

When spin-orbit coupling (SOC) is introduced in the Kagome lattice, it causes the Dirac points in the energy spectrum to be gapped out. However, due to the presence of time reversal symmetry, the formation of a Chern insulator phase with a non-zero Chern number is prohibited. On the other hand, if the system is prepared in a ferromagnetic state where all spins are aligned out of the plane, a $\mathcal{C} = -2$ Chern insulator phase can emerge [48]. This is because the broken time reversal symmetry allows for the development of a non-zero Chern number. This can be understood by considering that each K valley hosts two Dirac points, and there are two valleys in total.

To analyze the system's behavior, we use linear response theory to calculate the anomalous Hall conductance (AHC) curve as a function of the chemical potential. Figure 1.3 displays the effects of Rashba SOC and magnetism on the energy spectrum. These combined effects result in the appearance of a topological gap around the original Dirac points. By setting the chemical potential within this gap, which can be achieved through electrical gating in experiments, the system enters a Chern insulator phase with $\mathcal{C} = -2$, as confirmed by the AHC curve in panel (c) of Figure 1.3.

In-plane anomalous Hall effect

We now study a specific type of anomalous Hall effect that can be observed in a breathing Kagome lattice. Unlike the lattice geometry discussed previously, this system lacks the C_{2z} symmetry due to the trimerization of atoms. We assume the Kagome ferromagnets can be prepared with the magnetization lying completely in the $x - y$ plane, as is the case for Fe_3Sn , where this plane is the easy axis for magnetization. When Rashba spin-orbit coupling (SOC) is considered, the system holds a C_{3v} point group symmetry.

The in-plane anomalous Hall effect (IPAHE) can appear when the magnetic field is

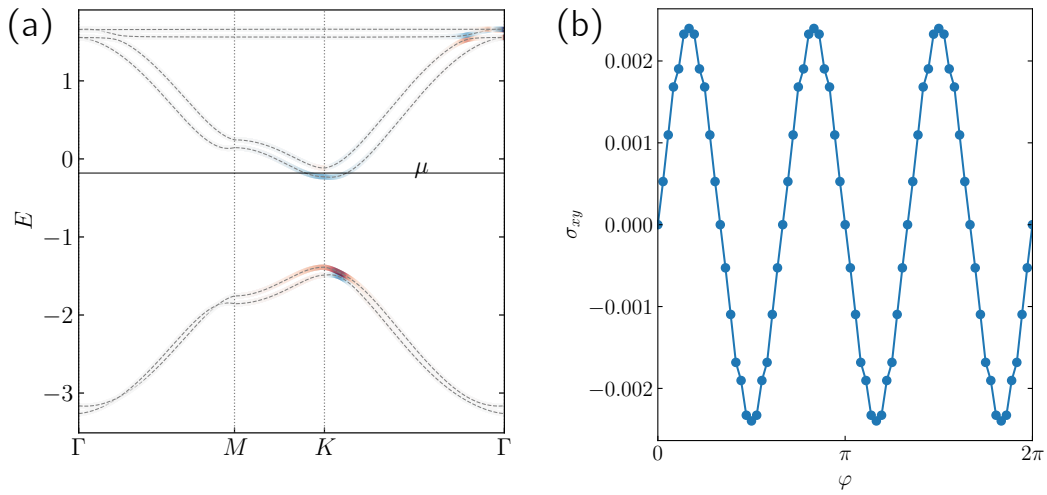


Figure 1.4: (a) Energy bands of a ferromagnetic Kagome lattice with Rashba-type spin-orbit coupling (SOC). The Rashba SOC opens a gap near the Dirac points, and the presence of magnetism breaks time reversal symmetry. The geometric setup is adapted from the structure of monolayer Fe_3Sn , which is a breathing Kagome lattice. The Berry curvature distribution along the high symmetry line is labeled in color. (b) The in-plane anomalous Hall response by changing the direction of the magnetic field. The Hall conductance σ_{xy} has a $2\pi/3$ periodicity which serve as a experimental signature of IPAHE. The chemical potential is set at μ which is labeled in the band structure plot.

confined to the $x - y$ plane, where the vertical mirror plane and $C_{2z}\mathcal{T}$ symmetry are broken. We first show in Fig. 1.4, the berry curvature has a non-trivial distribution in momentum space, where the SOC plays an important role. When the in-plane magnetic field is applied, the magnetization will align with this external field and φ describes the angle between the magnetization vector and the x -axis. Note that, for breathing Kagome ferromagnets, the system still has three mirror planes that are perpendicular to the $x - y$ plane, which results in nodes of the Hall conductance curve versus the angle φ , shown in Fig. 1.4. The anomalous Hall conductance σ_{xy} exhibits a $2\pi/3$ periodicity with respect to φ . This is a distinctive feature of IPAHE, which differs from the π periodicity observed for the case of an out-of-plane magnetic field. This difference provides a great advantage for experiments to distinguish the in-plane anomalous Hall effect from the normal Hall effect. This interesting IPAHE has been observed in Fe_3Sn_2 [49], while it is an antiferromagnetic material.

1.3.2 Moiré MoTe_2

Moiré materials, including twisted MoTe_2 , have recently been found to exhibit non-trivial band topology and host intriguing phenomena such as the quantum anomalous Hall effect (QAHE) and fractional quantum anomalous Hall effect (FQAHE) [1, 15, 16] without requiring an external magnetic field. These effects manifest as quantized Hall conductance at fractional values ($\nu = -2/3, -3/5$) and integer value ($\nu = -1$). The twist angle between the layers in MoTe_2 creates a moiré pattern that modifies the electronic band structure, leading to the emergence of topological states. This behavior arises from the interplay of the moiré superlattice potential, strong electron-electron interactions, and the intrinsic spin-orbit coupling in the material. The discovery of QAHE and FQAHE in twisted MoTe_2 (twist angle $\theta \approx 4^\circ$) opens up exciting prospects

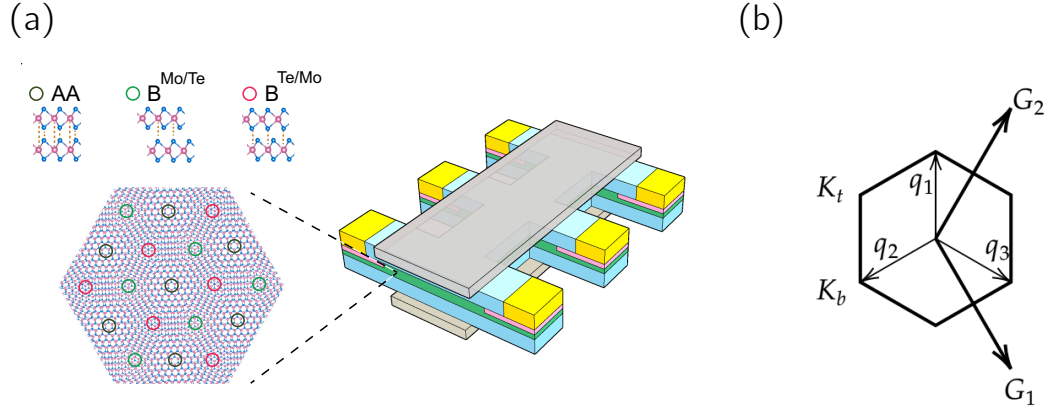


Figure 1.5: (a) Real space geometry of the twisted MoTe₂ and the experimental setup with a six-terminal Hall bar. This figure is taken from Ref. [1] under the terms of the Creative Commons Attribution 4.0 International license. (b) Geometry of the reciprocal lattice, where $\mathbf{G}_3 = -\mathbf{G}_1 - \mathbf{G}_2$, $\mathbf{G}_4 = \mathbf{G}_1 + \mathbf{G}_2$, $\mathbf{G}_5 = -\mathbf{G}_1$, $\mathbf{G}_6 = -\mathbf{G}_2$ and $\mathbf{q}_1 = \mathbf{K}_t - \mathbf{K}_b$.

for studying topological physics and developing innovative electronic devices based on moiré materials.

In this section, we will derive the continuum Hamiltonian [50] for twisted MoTe₂ (*t*MoTe₂) by considering symmetry constraints. This derivation will provide insights into the band topology of the material and its electronic properties. When the twist angle is small, two atomic valleys are well decoupled like the case of Bistritzer-Macdonald model for TBG [9]. The empirical continuum Hamiltonian for *K*-valley moiré electrons is written as,

$$H = \begin{pmatrix} -\frac{\hbar^2}{2m}(\mathbf{k} - \mathbf{K}_b)^2 + \Delta_b(\mathbf{r}) & \Delta_T(\mathbf{r}) \\ \Delta_T^\dagger(\mathbf{r}) & -\frac{\hbar^2}{2m}(\mathbf{k} - \mathbf{K}_t)^2 + \Delta_t(\mathbf{r}) \end{pmatrix} \quad (1.29)$$

The Hamiltonian for the *K'* valley in twisted MoTe₂ can be obtained by applying a time reversal operation. The diagonal part of the Hamiltonian describes a massive electron influenced by the intralayer moiré potential $\Delta_{t/b}(\mathbf{r})$. On the other hand, the

off-diagonal part $\Delta_T(\mathbf{r})$ represents the interlayer moiré potential induced by the twist. Indeed, the intralayer moiré potential in transition metal dichalcogenide (TMD) materials can often be significant due to the strong in-plane lattice reconstruction. This is in contrast to twisted bilayer graphene (TBG), where the intralayer moiré potential is typically weaker. We gauge out this Hamiltonian through a unitary transformation $UHU^\dagger$ to perform symmetry analysis, where

$$U = \begin{pmatrix} e^{-i\mathbf{K}_b \cdot \mathbf{r}} & 0 \\ 0 & e^{-i\mathbf{K}_t \cdot \mathbf{r}} \end{pmatrix}. \quad (1.30)$$

The transformed Hamiltonian is written as,

$$H(\mathbf{p} = \hbar\mathbf{k}, \mathbf{r}) = UHU^\dagger = \begin{pmatrix} -\frac{\hbar^2\mathbf{k}^2}{2m} + \Delta_b(\mathbf{r}) & e^{i(\mathbf{K}_t - \mathbf{K}_b) \cdot \mathbf{r}} \Delta_T(\mathbf{r}) \\ e^{-i(\mathbf{K}_t - \mathbf{K}_b) \cdot \mathbf{r}} \Delta_T^\dagger(\mathbf{r}) & -\frac{\hbar^2\mathbf{k}^2}{2m} + \Delta_t(\mathbf{r}) \end{pmatrix} \quad (1.31)$$

The transformed Hamiltonian should possess C_3 symmetry, satisfying the condition $C_3^{-1}H(\mathbf{p}, \mathbf{r})C_3 = H(C_3^{-1}\mathbf{p}, C_3^{-1}\mathbf{r})$. This symmetry requirement ensures that both $\Delta_{b/t}(\mathbf{r})$ and $e^{i(\mathbf{K}_t - \mathbf{K}_b) \cdot \mathbf{r}} \Delta_T(\mathbf{r})$ remain invariant under C_3 operations. Moreover, the moiré potential $\Delta(\mathbf{r})$ should exhibit moiré translational invariance, suggesting that it can be expressed as a series of plane waves associated with the moiré reciprocal lattice vectors $\mathbf{G}$. We can write down the expression for the moiré potential with first order of harmonics,

$$\begin{aligned} \Delta_{b/t}(\mathbf{r}) &= \sum_{\mathbf{G}} V_{\mathbf{G}} e^{i\mathbf{G} \cdot \mathbf{r}} = \sum_{j=4,5,6} 2V \cos(\mathbf{G}_j \cdot \mathbf{r} \pm \phi), \\ \Delta_T(\mathbf{r}) &= W(1 + e^{i\mathbf{G}_1 \cdot \mathbf{r}} + e^{-i\mathbf{G}_2 \cdot \mathbf{r}}). \end{aligned} \quad (1.32)$$

W, V are parameters describing the strength of the moiré potential and ϕ suggests the geometry of the intralayer one. We can further check the off diagonal part of the

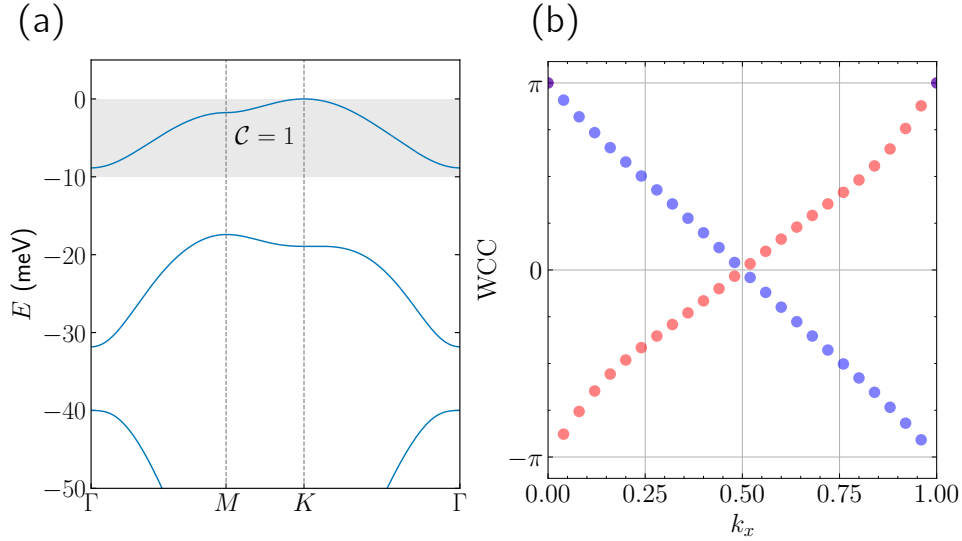


Figure 1.6: (a) Energy bands of twisted MoTe_2 with a twist angle of $\theta = 3.89^\circ$, obtained using an empirical continuum model at the K valley. The first valence band is remarkably flat, exhibiting a bandwidth of 10 meV, which is labeled in a light grey area. (b) Wannier Charge Center (WCC) flow for the two highest valence bands at the K valley which suggests they host a non-zero Chern number with $|\mathcal{C}| = 1$.

transformed Hamiltonian is C_3 invariant. Using the geometry depicted in Fig. 1.5, we have,

$$e^{i(\mathbf{K}_t - \mathbf{K}_b) \cdot \mathbf{r}} \Delta_T(\mathbf{r}) = e^{i\mathbf{q}_1 \cdot \mathbf{r}} (1 + e^{i\mathbf{G}_1 \cdot \mathbf{r}} + e^{-i\mathbf{G}_2 \cdot \mathbf{r}}) = e^{i\mathbf{q}_1 \cdot \mathbf{r}} + e^{i\mathbf{q}_2 \cdot \mathbf{r}} + e^{i\mathbf{q}_3 \cdot \mathbf{r}}, \quad (1.33)$$

and of course this term is invariant under C_3 rotation. The parameters for the continuum Hamiltonian are obtained through fitting to first principles calculations, which can be a subject of controversy since different parameter sets can lead to different topological behaviors of the bands [50, 51]. We adopt the parameters from Ref. [51] ($W = -23.8 \text{ meV}$, $V = 20.8 \text{ meV}$, $\phi = 107.7^\circ$, $m = 0.6m_e$). This parameter set has been subsequently confirmed by another large-scale first principles calculation [52].

The single-particle Hamiltonian for the K valley can be solved using a plane wave

expansion, as illustrated in Figure 1.6-(a). Notably, the top valence band exhibits an exceptionally flat shape, with a narrow bandwidth of approximately 10 meV, and it is well-separated from the other bands. This observation suggests that the electrons in the valence bands can exhibit strong correlation effects, resembling a moiré Hubbard model.

Additionally, the Wannier Charge Center (WCC) flow depicted in Figure 1.6-(b) indicates that both of the two highest bands possess a non-zero Chern number of $|\mathcal{C}| = 1$. This intriguing topological behavior, combined with the flat nature of the bands, creates the potential for the Quantum Anomalous Hall (QAH) effect to emerge. This effect can manifest when the bands become valley-polarized and the time reversal symmetry is spontaneously broken as a result of strong electron-electron interactions.

1.4 Structure of the dissertation

With recent experimental progress in moiré materials, an increasing number of intriguing phenomena have been observed, as discussed earlier. However, the ground state of these materials has remained a topic of debate for a significant period. The large size of the superlattice in moiré materials makes it computationally expensive to perform first-principles calculations and construct accurate low-energy models at the single-particle level. Consequently, gaining a precise understanding of the many-particle behavior in moiré materials has proven challenging. From an experimental perspective, researchers are interested in exploring whether similar extraordinary phenomena can be discovered in a wider range of materials, guided by insights from theoretical studies. Therefore, researchers are now considering the possibility of introducing moiré effects in traditional semiconductors. This approach would involve

leveraging techniques such as gate engineering and precise growth methods to control various properties in these materials. This motivation has prompted theorists to investigate the concept of artificial moiré lattices without the need for a twist.

In this dissertation, a novel method for accurately capturing the low-energy Hamiltonian of moiré materials is proposed and tested. Then we focus on the moiré effects on thin films of Cd_3As_2 . Here is the brief summary for this dissertation,

Chapter 2: Introduction to the Truncated Atomic Plane Wave (TAPW) method for moiré materials.

Chapter 3: Study of moiré phonons and electron optical phonon coupling in MATBG.

Chapter 4: Investigation of magneto transport phenomena in molecular beam epitaxy (MBE)-grown Cd_3As_2 thin films.

Chapter 5: Exploration of the artificial moiré lattices and the relevant band topology.

Chapter 6: Summary of the dissertation and outlook of the future directions.

Chapter 2

Truncated atomic plane wave method for subband structure calculations in moiré systems

We propose a highly efficient and accurate numerical scheme named the truncated atomic plane wave (TAPW) method to determine the subband structure of twisted bilayer graphene (TBG) inspired by the Bistritzer-MacDonald model. Our method utilizes real-space information of carbon atoms in the moiré unit cell and projects the full tight-binding Hamiltonian into a much smaller subspace using atomic plane waves. Using our method, we are able to present accurate electronic band structures of TBG in a wide range of twist angles. The higher order Fourier components of the moiré potential and non-local tunneling effect can be naturally captured in this method.

The contents of this chapter have substantially appeared in Ref. [53]. This article is copyrighted by the American Physical Society and reproduced with permission.

2.1 Introduction

Twisted Bilayer Graphene (TBG), formed by stacking one single layer on the other with a small twist, provides a great platform for physicists to study novel quantum phenomena. More attention has been attracted after the discovery of unconventional superconductivity, orbital magnetism and correlated insulating phases [54–71] in TBG systems at the magic angle, around 1.1° . The mechanism behind these observations is still an open question.

Unlike aligned bilayer graphene, in TBG systems, a moiré pattern forms in the real space which breaks original periodicity of graphene. This pattern leads to the difficulty for band structure calculations due to the huge amount of atoms in a single unit cell. In order to overcome this kind of difficulty, several low energy effective models [9, 72–83] were developed, by which the most interesting physics in TBG systems at the magic angle – flat bands, has been predicted theoretically. In the widely used Bistritzer-MacDonald (BM) model [9], the moiré potential is expanded to leading order by Fourier Transformation and a general model Hamiltonian for arbitrary twist angles can be constructed. With such model, B&M predict that flat bands emerge when twist angle is around 1.1° which is called “magic angle”. The “magic” happens because the low energy band is so flat that the velocity of electrons vanishes. In these flat band systems, the electron-electron interactions dominate the band structure and could result in various exotic correlated phenomena. More accurate calculation based on density functional theory (DFT) [84–87] can also be performed using large scale parallelized DFT code integrated with van der Waals functional to catch the details of the band structure. However, this kind of computation is very time consuming and hard to implement for systems with small twist angles.

Although BM model provides a clear picture of flat bands near the first magic angle and is easy to implement numerically, the model itself ignores microscopic details such as atomic relaxation [76, 84–86, 88–94] in the moiré scale which can explain the insulating phase of MATBG at ± 4 filling. For many 2D materials, tight binding (TB) models are often used to describe electronic band structures. Compared with BM model, tight binding scheme can easily take atomic relaxation into consideration by resetting the coordinates of carbon atoms and the related hopping parameters can be determined by fitting to small scale DFT calculation results. Full tight binding calculation [95–99] in the framework of Slater-Koster theory [17] is performed and provides reliable results. Several *ab initio* TB models [81–83, 100–102] have been carefully designed to make the band structures of TBG much closer to DFT results. However, full TB model will generate a huge Hamiltonian matrix when twist angle is small which makes further calculations hard to perform.

In this manuscript, we present a well designed numerical scheme named Truncated Atomic Plane Wave (TAPW) method to project the full tight binding Hamiltonian onto the truncated atomic plane waves. TAPW shows very accurate band structures compared with those retrieved from full TB Hamiltonian. The key point of our new method is to combine the advantages of both the BM model and full TB model. First, like full TB model, the atomic plane wave basis will be constructed with full information of the real atomic positions. Next, like the treatment in BM model, for small enough twist angles, the atomic valley degree of freedom can still be treated as a good quantum number and then the complete Hilbert space of full TB model can be truncated into two groups of atomic plane waves, where only the wave vectors close enough to the graphene $\mathbf{K}/\mathbf{K}'$ points are included. After the basis truncation, the total dimension of the Hamiltonian can be reduced significantly from several tens of thousand to only

several hundreds. Our scheme has the following advantages compared with existing models:

1. It saves computing resources compared with a full TB scheme but shares the same accuracy of low energy bands.
2. The parameters of the TB model can be carefully designed, using a Slater Koster scheme [95–99] or an *ab initio* TB scheme [81–83, 100–102].
3. The whole workflow is simple and straightforward without manually expanding BM model to higher orders [76, 79, 80, 82, 83], or fitting parameters [80, 103].
4. Follow up studies, such as Hartree Fock (HF), constrained Random Phase Approximation (cRPA) or dynamic mean field (DMFT) calculations can be carried out based on our method.
5. The relaxation effect and symmetry constraint can be considered appropriately by setting real space coordinates of carbon atoms like other TB models.

Based on this kind of numerical strategy, we develop an open source Python package hosted on GitHub ¹. We also extend our method to describe the moiré phonons and determine the screened Coulomb interaction of Magic Angle Twisted Bilayer Graphene (MATBG) using constrained Random Phase Approximation (cRPA).

¹[GitHub Repository Link for TAPW](#)

2.2 From BM model to Truncated Atomic Plane Wave method

2.2.1 Geometry of TBG

We define $\mathbf{a}_1 = a(\sqrt{3}/2, -1/2)$, $\mathbf{a}_2 = a(\sqrt{3}/2, 1/2)$ as the lattice vectors for the atomic structure of monolayer graphene and $\mathbf{b}_1 = 2\pi/a(\sqrt{3}/3, -1)$, $\mathbf{b}_2 = 2\pi/a(\sqrt{3}/3, 1)$ as their corresponding reciprocal lattice vectors. $a = 0.246$ nm is the graphene lattice constant. The geometry of TBG can be defined by rotating two different layers of AA-stacking bilayer graphene around the AA-stacking point. After anti-clockwisely rotating the layer (1) by $+\theta/2$ and the layer (2) by $-\theta/2$, the lattice vectors should be $\mathbf{a}_i^{(1)} = D\left(\frac{\theta}{2}\right)\mathbf{a}_i$, $\mathbf{a}_i^{(2)} = D\left(-\frac{\theta}{2}\right)\mathbf{a}_i$ and the reciprocal lattice vectors are $\mathbf{b}_i^{(1)} = D\left(\frac{\theta}{2}\right)\mathbf{b}_i$, $\mathbf{b}_i^{(2)} = D\left(-\frac{\theta}{2}\right)\mathbf{b}_i$, where $D(\theta)$ is a 2D rotation matrix.

TBG system will have a commensurate structure [72, 73, 104] if the twist angle obeys:

$$\theta = \arcsin\left(\frac{\sqrt{3}(2N+1)}{6N^2+6N+2}\right), \quad (2.1)$$

where N is an integer. As illustrated in Fig.2.1, the unit lattice vectors for the moiré super cell are:

$$\begin{aligned} \mathbf{L}_1 &= D\left(\frac{\theta}{2}\right)[-N\mathbf{a}_1 + (2N+1)\mathbf{a}_2], \\ \mathbf{L}_2 &= D\left(\frac{\theta}{2}\right[-(2N+1)\mathbf{a}_1 + (N+1)\mathbf{a}_2]. \end{aligned} \quad (2.2)$$

The corresponding moiré reciprocal lattice vectors can be chosen as: $\mathbf{G}_1 = \mathbf{b}_1^{(1)} - \mathbf{b}_1^{(2)}$, $\mathbf{G}_2 = \mathbf{b}_2^{(1)} - \mathbf{b}_2^{(2)}$. The system has a D_3 point group symmetry².

²Systems with a D_6 point group symmetry are also provided in our Python Packages.

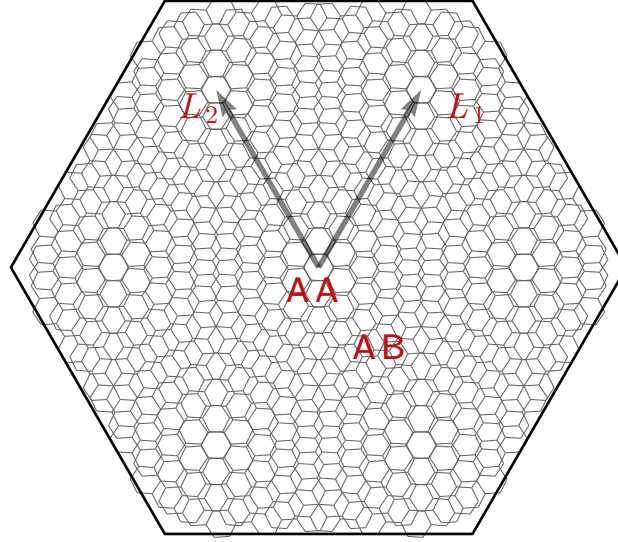


Figure 2.1: Schematic diagram for the real space twist using the geometry stated in the main context when $N = 4$, i.e. $\theta = 7.341^\circ$. $\mathbf{L}_1, \mathbf{L}_2$ are moiré unit lattice vectors. Corresponding $\mathbf{k}$ space diagram is shown in Fig. 2.2. AA stacking region and AB stacking region are also labeled.

2.2.2 BM Model

A widely used way to deal with the band structure of TBG at small twist angle is BM model [9, 72–75]. In BM model, the Hamiltonian of valley $\xi = \pm 1$ written in real space is:

$$H_\xi(\mathbf{r}) = \begin{pmatrix} h^{(1)}(\mathbf{r}) & U(\mathbf{r}) \\ U^\dagger(\mathbf{r}) & h^{(2)}(\mathbf{r}) \end{pmatrix}, \quad (2.3)$$

where $h(\mathbf{r})$ represents for intralayer part of the Hamiltonian and $U(\mathbf{r})$ is the large scale moiré potential. The analysis for BM can be done in $\mathbf{k}$ -space. For a two-layer system

like TBG, the Bloch basis for each layer (or atomic Bloch basis) can be defined as:

$$\begin{aligned}\psi_{\mathbf{k},\alpha}^{(1)}(\mathbf{r}) &= \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_\alpha^{(1)}} e^{i\mathbf{k}\cdot\mathbf{R}_\alpha^{(1)}} \phi_{p_z}(\mathbf{r} - \mathbf{R}_\alpha^{(1)}), \\ \psi_{\mathbf{p},\beta}^{(2)}(\mathbf{r}) &= \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_\beta^{(2)}} e^{i\mathbf{p}\cdot\mathbf{R}_\beta^{(2)}} \phi_{p_z}(\mathbf{r} - \mathbf{R}_\beta^{(2)}),\end{aligned}\tag{2.4}$$

where N is the number of atomic unit cell in each layer, and $\mathbf{R}_\alpha, \mathbf{R}_\beta$ is the concrete position of A/B carbon atoms, (1)/(2) is a notation for different layers.

Under this kind of basis function, $h(\mathbf{r})$ is naturally diagonalized with $\mathbf{k}$ and can be approximated as a Dirac equation when $\mathbf{k}$ or $\mathbf{p}$ is near atomic $\mathbf{K}, \mathbf{K}'$ points. However, $U(\mathbf{r})$ will couple different $\mathbf{k}$ and $\mathbf{p}$ satisfying the following condition [74]:

$$\mathbf{k} = \mathbf{p} - m_1 \mathbf{G}_1 - m_2 \mathbf{G}_2,\tag{2.5}$$

m_1, m_2 are two integers. BM approximates $U(\mathbf{r})$ by keeping three leading Fourier components:

$$\begin{aligned}U(\mathbf{r}) &= \begin{pmatrix} U_{A_1A_2} & U_{A_1B_2} \\ U_{B_1A_2} & U_{B_1B_2} \end{pmatrix} \\ &= \begin{pmatrix} u & u' \\ u' & u \end{pmatrix} + \begin{pmatrix} u & u'\omega^\xi \\ u'\omega^{-\xi} & u \end{pmatrix} e^{i\xi(-\mathbf{G}_1)\cdot\mathbf{r}} + \begin{pmatrix} u & u'\omega^{-\xi} \\ u'\omega^\xi & u \end{pmatrix} e^{i\xi\mathbf{G}_2\cdot\mathbf{r}},\end{aligned}\tag{2.6}$$

where $\omega = e^{(i2\pi/3)}$, and we adopt $u = 0.08581 \text{ eV}, u' = 0.1032 \text{ eV}$ after considering corrugation effect. Parameters are retrieved from our new method, see Appx. A.2.

2.2.3 Truncated Atomic Plane Wave method: theoretical formalism

The BM model is a good approximation for low energy physics of TBG at the first magic angle. However, band structures solved from BM model are not reliable any more when twist angle gets smaller where relaxation effect plays an important role [80, 88, 89, 92–94]. Parameters like u, u' in the BM model are highly sensitive to the structure of TBG and more Fourier components of $U(\mathbf{r})$ should be considered. To develop a more accurate description for the low energy physics of TBG, we can still follow the main idea and keep the key approximation of BM model:

1. Expand the Hamiltonian using atomic Bloch function defined in Eq.(2.4), see Ref. [9, 74–76, 79].
2. Intervalley tunneling process can be safely ignored at small twist angle, so the system has an approximate $U_v(1)$ symmetry, see Ref. [9, 72, 80, 81].
3. Since the low energy physics mainly comes from atomic $\mathbf{K}$ or $\mathbf{K}'$ points, only a group of plane waves close to Dirac points should be taken into consideration, see Ref. [9, 75, 76, 79, 82, 83].

These observations from the BM model studies first inspire us to directly expand the Hamiltonian of TBG utilizing atomic Bloch basis defined in Eq.(2.4). When the system is commensurate, the atomic Bloch basis could be modified to match the moiré superlattice. More concretely, the following substitution should work

$$N = N_m N_a, \quad \mathbf{k} = \bar{\mathbf{k}} + \mathbf{G}_n, \quad (2.7)$$

where N_m is the number of moiré lattices and $N_a = M/4$, M is the number of all atoms in a moiré superlattice. $\bar{\mathbf{k}}$ is defined in the first moiré B.Z. and $\mathbf{G}_n$ is the moiré recip-

rocal lattice vector. These substitution transforms the atomic Bloch function, which is Eq.(2.4), to

$$|\psi_{\alpha n}(\bar{\mathbf{k}})\rangle = \frac{1}{\sqrt{N_m N_a}} \sum_{I,i} e^{i(\bar{\mathbf{k}} + \mathbf{G}_n) \mathbf{R}_{Ii\alpha}} |\phi_{p_z}(\mathbf{r} - \mathbf{R}_{Ii\alpha})\rangle, \quad (2.8)$$

where $\mathbf{R}_{Ii\alpha} = \mathbf{L}_I + \boldsymbol{\tau}_{i\alpha}$ for short and $\mathbf{L}_I$ is the lattice vector of the moiré unit cell. $\alpha = A_1, B_1, A_2, B_2$ denotes the sublattice, $\boldsymbol{\tau}_{i\alpha}$ is the displacement of atom α in the i -th atomic cell with respect to the I -th moiré cell. The basis wavefunction, which we call *atomic plane wave basis*, can be viewed as a Bloch summation of atomic p_z orbitals, and for unrelaxed TBG, it is naturally normalized.

After using atomic plane wave basis defined in Eq.(2.8) to expand the Hamiltonian and further taking advantage of Eq.(2.5), we can write down the matrix element $H_{\alpha n, \beta m}$ of the Hamiltonian:

$$\begin{aligned} & \langle \psi_{\alpha n}(\bar{\mathbf{k}}) | \hat{H} | \psi_{\beta m}(\bar{\mathbf{k}}) \rangle \\ &= \frac{1}{N_m N_a} \sum_{I,J,i,j} t(\mathbf{R}_{Ii\alpha} - \mathbf{R}_{Jj\beta}) e^{-i(\bar{\mathbf{k}} + \mathbf{G}_n) \mathbf{R}_{Ii\alpha}} e^{i(\bar{\mathbf{k}} + \mathbf{G}_m) \mathbf{R}_{Jj\beta}} \\ &= \frac{1}{N_m N_a} \sum_{I,J,i,j} e^{-i\mathbf{G}_n \boldsymbol{\tau}_{i\alpha}} e^{-i\bar{\mathbf{k}}(\mathbf{L}_I - \mathbf{L}_J + \boldsymbol{\tau}_{i\alpha} - \boldsymbol{\tau}_{j\beta})} t(\mathbf{L}_I - \mathbf{L}_J + \boldsymbol{\tau}_{i\alpha} - \boldsymbol{\tau}_{j\beta}) e^{i\mathbf{G}_m \boldsymbol{\tau}_{j\beta}} \\ &= \frac{1}{N_m N_a} \sum_{I,i,j} e^{-i\mathbf{G}_n \boldsymbol{\tau}_{i\alpha}} e^{-i\bar{\mathbf{k}}(\bar{\boldsymbol{\tau}}_{i\alpha,j\beta})} t(\bar{\boldsymbol{\tau}}_{i\alpha,j\beta}) e^{i\mathbf{G}_m \boldsymbol{\tau}_{j\beta}} \\ &= \sum_{ij} \left(\frac{e^{-i\mathbf{G}_n \boldsymbol{\tau}_{i\alpha}}}{\sqrt{N_a}} \right) \cdot \left[e^{-i\bar{\mathbf{k}}(\bar{\boldsymbol{\tau}}_{i\alpha,j\beta})} t(\bar{\boldsymbol{\tau}}_{i\alpha,j\beta}) \right] \cdot \left(\frac{e^{i\mathbf{G}_m \boldsymbol{\tau}_{j\beta}}}{\sqrt{N_a}} \right). \end{aligned} \quad (2.9)$$

I, J are moiré lattice indices and $i\alpha, j\beta$ are position indices for carbon atoms in the moiré superlattice. α, β runs over $[A_1, B_1, A_2, B_2]$. $\bar{\boldsymbol{\tau}}_{i\alpha,j\beta}$ is the distance between atom $i\alpha$ and atom $j\beta$. $t(\bar{\boldsymbol{\tau}}_{i\alpha,j\beta})$ is the hopping integral under tight binding approximation. Furthermore, the matrix form of the Hamiltonian, which we denote as $\mathbf{H}^{\text{TAPW}}$, can be

written in a more compact way by inspecting the last step of Eq.(2.9)

$$\begin{aligned}\mathbf{H}^{\text{TAPW}} &= \sum_{\alpha\beta} \mathbf{X}_{\alpha}^{\dagger} \mathbf{T}_{\alpha\beta} \mathbf{X}_{\beta} \\ &= \mathbf{X}^{\dagger} \mathbf{T} \mathbf{X},\end{aligned}\tag{2.10}$$

where the corresponding matrix elements are

$$\begin{aligned}(\mathbf{X}_{\alpha})_{n,i} &= e^{i\mathbf{G}_n \cdot \mathbf{r}_{i\alpha}} / \sqrt{N_a}, \\ (\mathbf{T}_{1\alpha\beta})_{i,j} &= e^{-i\bar{\mathbf{k}} \cdot (\bar{\mathbf{r}}_{i\alpha,j\beta})}, \\ (\mathbf{T}_{2\alpha\beta})_{i,j} &= t(\bar{\mathbf{r}}_{i\alpha,j\beta}).\end{aligned}\tag{2.11}$$

$\mathbf{T} = \mathbf{T}_1 * \mathbf{T}_2$ and “*” is an element-wise product.

In Eq.(2.10), $\mathbf{T}$ is exactly the full tight binding matrix and $\mathbf{X}$ is a plane wave projector under continuum approximation $e^{i\mathbf{G}_n \cdot \mathbf{r}_{i\alpha}} = e^{i\mathbf{G}_n \cdot \mathbf{r}} \delta(\mathbf{r} - \mathbf{r}_{i\alpha}) \approx e^{i\mathbf{G}_n \cdot \mathbf{r}}$. When $\mathbf{G}$ runs over all the moiré reciprocal lattice vectors in the graphene first B.Z., Eq.(2.10) is a unitary transformation and it will restore the full TB model. Following the spirit of the BM model, we do *truncation* on the $\mathbf{G}$ vectors within certain fixed distance away from graphene $\mathbf{K}/\mathbf{K}'$ points when calculating the band structure of TBG at a small twist angle, as illustrated in Fig.2.2. We want to further emphasize, our method is intrinsically equivalent to the generalized BM model because we start from the same basis function (atomic Bloch function) to expand the Hamiltonian. However, our method requires the system to be commensurate, then Eq.(2.8) can be established. The advantage of our new method is that TAPW directly projects the full TB matrix on a series of plane waves without assuming the tunneling amplitude t between different layers is a smooth function and then manually performing Fourier expansion [9, 74–76, 79, 80, 82, 83]. Interestingly, our TAPW method presents an exact mapping between the full TB Hamil-

tonian and generalized continuum model, which is a simple realization of Ref. [77].

For a better understanding of the superiority of our TAPW method compared with the full TB model, we first introduce some numerical details in our realization. Note that the size of $\mathbf{G}$ -list is N_G and the number of atoms in the moiré superlattice is $M = 4 \times N_a$. The dimension of the Hamiltonian matrix $\mathbf{H}$ is $4N_G \times 4N_G$, the tight binding matrix $\mathbf{T}$ is $M \times M$ and the plane wave projection matrix $\mathbf{X}$ is $4N_G \times M$. Our TAPW method projects the sparse TB matrix $\mathbf{T}$ into a much smaller subspace. Typically for MATBG ($\theta = 1.085^\circ$, $M = 11164$), we can restore electronic band structure near Fermi level (± 0.6 eV) perfectly compared with full TB result using only 244 plane waves per valley, i.e., $N_G = 61$. The TB Hamiltonian for each $\mathbf{k}$ point is downfolded from 11164×11164 to 244×244 per valley. We transform the computational complexity of diagonalizing a huge sparse matrix into the multiplication of sparse matrices together with diagonalizing a much smaller dense one. The latter operation saves a huge amount of computational power and preserves low energy electronic band structure (see detailed discussion in Sec. 2.3). The sparse matrix operations are boosted by SciPy [105]. It is worth pointing out the setup of $\mathbf{X}$, $\mathbf{T}_2$ is only once during the whole computational process because they are not $\bar{\mathbf{k}}$ dependent.

The construction of plane wave projection matrix $\mathbf{X}$ is fast and the corresponding computational complexity is $\mathcal{O}(M \times N_G)$. However, setting up transfer integral matrix $\mathbf{T}_2$ and hopping phase matrix $\mathbf{T}_1$ is very time consuming because we have to determine the neighbours of a specific carbon atom in such a large system with more than 10,000 atoms. The brute force searching scheme has a complexity of $\mathcal{O}(M^2)$ and it fails full TB model in dealing with a much smaller twist angle.

Another strategy we adopt is using k -d tree [106] to optimize the searching scheme of determining neighbours. We utilize the fact there are only carbon atoms in the TBG

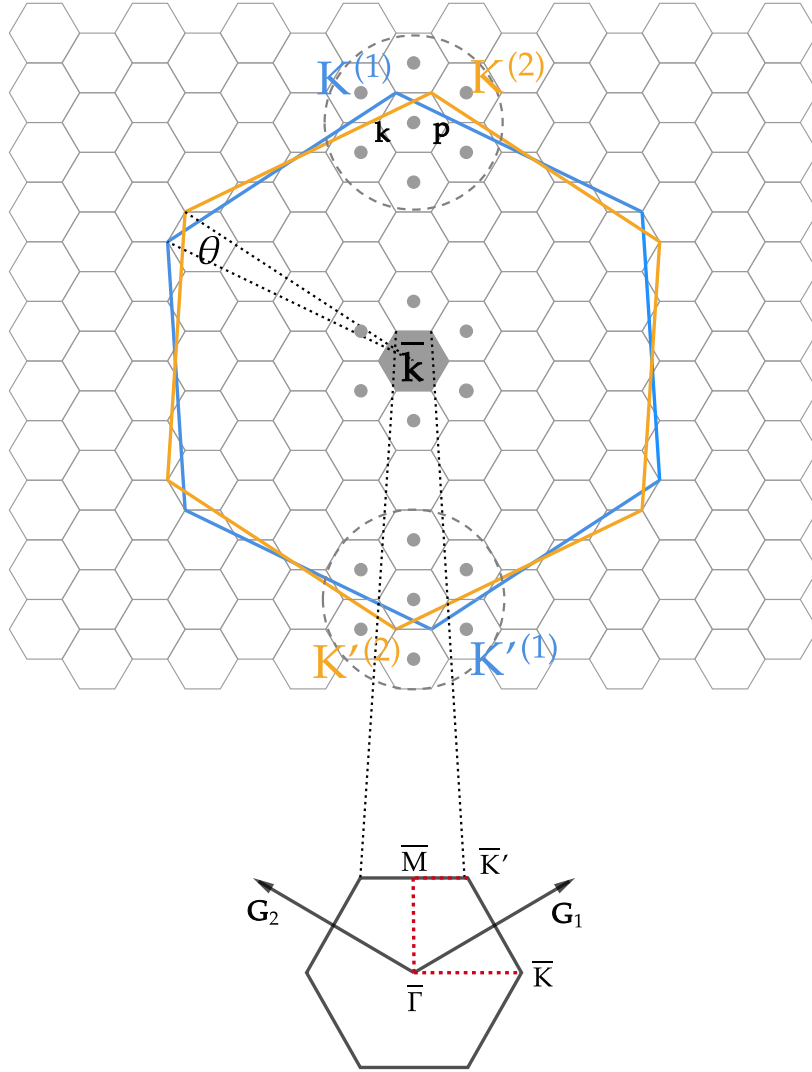


Figure 2.2: Schematic diagram for rotation in the k space for $N = 4$, i.e. $\theta = 7.341^\circ$. We construct $\mathbf{G}$ -list in two circled areas (centered around $\mathbf{K}, \mathbf{K}'$) for two different valleys when calculating the band structure for moiré electrons and are moiré reciprocal lattice vector. The small grey dots represent the plane waves used to expand the TB Hamiltonian with the first shell conserved. The band structures of TBG are calculated along high symmetry path (denoted in a red dashed line).

system and the hopping process can mainly happen between one specific carbon atom and another from the nearest moiré unit cell. Thus, we construct a 3×3 super cell as the searching space. Such super cell may consist of millions of carbon atoms but it only consumes a small amount of memory space to store 3D coordinates. As demonstrated in Fig.2.3-(a), a highly efficient k -d tree searching scheme is applied which reduces the related computational complexity to $\mathcal{O}(M \log M)$ and $t(\bar{\tau}_{i\alpha,j\beta})$ is calculated in $\mathcal{O}(1)$ time enhanced by the vectorization characteristic of NumPy [107].

2.2.4 Corrugation and relaxation

In TAPW, the information of atomic position in the moiré scale is encoded through tight binding description by definition. The deviation of TBG system from a rigid structure can be considered naturally by setting the 3D coordinates of carbon atoms. As observed in the DFT calculation [84–86] and molecular dynamics simulations [76, 92–94] for TBG near the first magic angle, the two graphene sheets are not totally flat and there exists some fluctuation in the real space. The z direction displacement can help separate flat bands away from remote bands near the first magic angle and further stabilize the insulating phase of MATBG at ± 4 filling [85]. This kind of corrugation effect can be easily simulated in our TAPW method by adding displacements along z direction for carbon atoms [75, 84],

$$\begin{aligned} d^{(1)} &= \frac{1}{2}d_0 + d_1 \sum_{n=1,2,3} \cos(\mathbf{G}_n \cdot \boldsymbol{\tau}), \\ d^{(2)} &= \frac{1}{2}d_0 - d_1 \sum_{n=1,2,3} \cos(\mathbf{G}_n \cdot \boldsymbol{\tau}), \end{aligned} \tag{2.12}$$

where $d_0 = 3.43 \text{ \AA}$ is average interlayer distance for TBG, $d_1 = 0.278 \text{ \AA}$ is obtained from looking at the difference of the interlayer distance between AA-stacking bilayer and

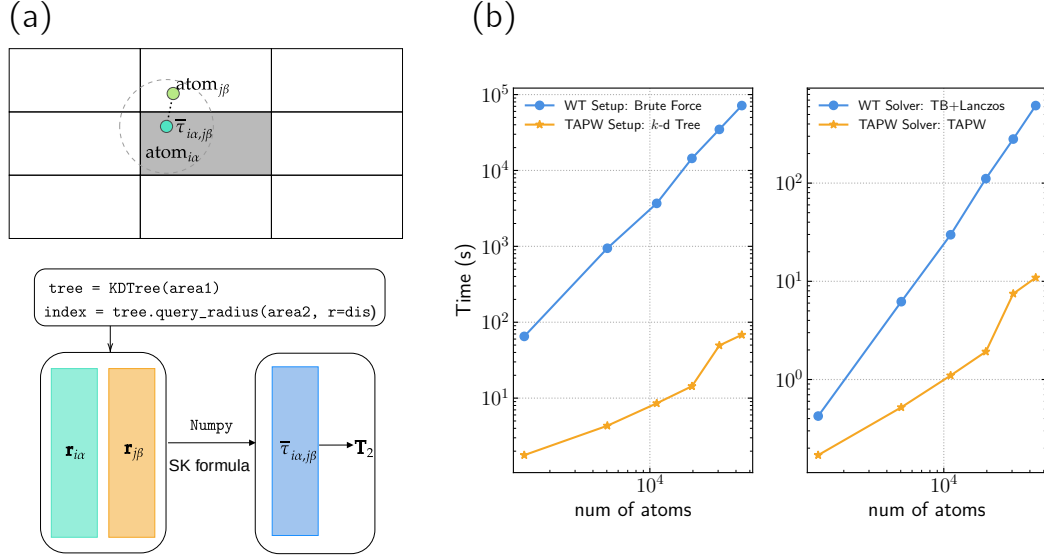


Figure 2.3: (a) We design an efficient numerical scheme to build the hopping matrix $\mathbf{T}_2$. The grey block represents for the moiré super cell and we set up a 3×3 super cell to perform a k -d tree search. The grey block is labeled as area2 and the larger one as area1. The searching process is performed using k -d tree algorithm integrated in sklearn [2] which returns the neighbour pair indices $(i\alpha, j\beta)$. The 3D coordinates for neighbour pairs $\mathbf{r}_{i\alpha} - \mathbf{r}_{j\beta}$ are stored in NumPy ndarray denoted with different color blocks. The computation for hopping parameter $\bar{\tau}_{i\alpha,j\beta}(\mathbf{r}_{i\alpha} - \mathbf{r}_{j\beta})$ is finished in $\mathcal{O}(1)$ time taking advantage of the built in vectorization mechanism of NumPy. Then the hopping matrix $\mathbf{T}_2$ can be constructed with neighbour pair indices and $\bar{\tau}$ array. (b) Computational efficiency comparison between full tight binding scheme and truncated atomic plane wave scheme. Tight binding solver for twisted bilayer graphene is a new feature of WannierTools (WT), a popular Fortran routine to solve tight binding Hamiltonian and related topological properties. TAPW is a submodule of our Python package. The left panel plots the time consumed to set up the tight binding kernel versus number of atoms in the moiré superlattice. WT uses a brute force searching scheme to build SK tight binding kernel while TAPW uses a k -d tree searching scheme. The right panel plots the time consumed to diagonalize the Hamiltonian per k point versus the number of atoms in the moiré superlattice. WT utilizes the power of ARPACK (Lanczos algorithm) to calculate the eigenvalues of the sparse tight binding Hamiltonian and TAPW uses the proposed projection algorithm. The benchmark is performed serially on an 8-core, 16-thread Intel Xeon W-3223 processor with WannierTools V2.6.2.

AB-stacking bilayer. $\mathbf{G}_3 = -(\mathbf{G}_1 + \mathbf{G}_2)$ is the third smallest moiré reciprocal lattice vector. $\boldsymbol{\tau}$ is the atomic position of the carbon atom in the moiré unit cell.

Moreover, the local geometry of rigid TBG can be classified into three different regions: AA-stacking area, AB/BA-stacking area and saddle point (SP) area. As pointed out in DFT study [92], AB/BA-stacking area and SP area are more energetically favorable compared with AA-stacking area, but the in-plane strain field can also compete with such kind of interlayer energy minimization. It is also evidenced in STM experiments [59], when the twist angle is small, TBG undergoes a self-organized lattice reconstruction to shrink AA-stacking area and expand AB-stacking area which forms a triangular lattice in the moiré scale. These microscopic in-plane distortions can be well captured in TAPW by reconstructing the Bloch function [79]:

$$\begin{aligned}\psi_{\mathbf{k},\alpha}^{(1)}(\mathbf{r}) &= \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_\alpha^{(1)}} e^{i\mathbf{k} \cdot \mathbf{R}_\alpha^{(1)}} \phi_{p_z}(\mathbf{r} - \mathbf{R}_\alpha^{(1)} - \mathbf{u}(\mathbf{R}_\alpha^{(1)})), \\ \psi_{\mathbf{p},\beta}^{(2)}(\mathbf{r}) &= \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_\beta^{(2)}} e^{i\mathbf{p} \cdot \mathbf{R}_\beta^{(2)}} \phi_{p_z}(\mathbf{r} - \mathbf{R}_\beta^{(2)} - \mathbf{u}(\mathbf{R}_\beta^{(2)})).\end{aligned}\tag{2.13}$$

$\mathbf{u}$ is an abstract displacement vector field which slowly varies in the atomic scale with a moiré periodicity. We can still follow the proposed procedure to perform calculation and take valley as a good quantum number when the twist angle is small. The Hamiltonian can be solved in an elegant way by just resetting hopping matrix $\mathbf{T}_2$ with the relaxed coordinates of the carbon atoms. The full atomic relaxation can be performed using classical molecular dynamics with the rigid structure as a start point.

2.3 Numerical results and comparison

In our numerics, the hopping integral $t(\bar{\tau}_{i\alpha,j\beta})$ is determined by Slater-Koster (SK-TB) formula [75, 98] or environment adapted *ab initio* tight binding parameters (FK-TB) [82, 83, 100, 101]. It is worthwhile to mention these are all based on two center approxiamtions.

For Slater-Koster (SK) formula, we set

$$t(\mathbf{r}) = -V_\pi \left(1 - \frac{r_z^2}{r^2}\right) - V_\sigma \frac{r_z^2}{r^2}, \quad (2.14)$$

with $r_z = \mathbf{r} \cdot \mathbf{e}_z$, $V_\pi = V_\pi^0 e^{-(r-a_0)/r_0}$, $V_\sigma = V_\sigma^0 e^{-(r-d_0)/r_0}$. where $d_0 = 0.335 \text{ nm}$ is the average interlayer spacing, $a_0 = a/\sqrt{3}$ is the nearest neighbour distance, $r_0 = 0.184a$ is the characteristic length of the hopping strength, the hopping amplitudes are set as $V_\pi^0 = -2.7 \text{ eV}$ and $V_\sigma^0 = 0.48 \text{ eV}$.

For FK-TB model, which is more accurate according to recent DFT research [101], the intra-layer hopping integral t_{intra} is determined by [82, 83]

$$t_{\text{intra}}(\mathbf{r}) = t_0 e^{-\alpha_0 \bar{r}^2} \cos(\beta_0 \bar{r}) + t_1 \bar{r}^2 e^{-\alpha_1 (\bar{r}-r_1)^2}. \quad (2.15)$$

Different from Slater-Koster description for inter-layer tunneling, which is isotropic, the inter-layer hopping integral $t_{\text{inter}}(\mathbf{r})$ in FK model is determined by [82, 83, 100, 101]

$$\begin{aligned} t_{\text{inter}}(\mathbf{r}) = & V_0(r) + V_3(r) \left(\frac{1}{3} \sum_{\alpha=1}^3 \cos(3\theta_{12}^{(\alpha)}) + \frac{1}{3} \sum_{\alpha=1}^3 \cos(3\theta_{21}^{(\alpha)}) \right) \\ & + V_6(r) \left(\frac{1}{3} \sum_{\alpha=1}^3 \cos(6\theta_{12}^{(\alpha)}) + \frac{1}{3} \sum_{\alpha=1}^3 \cos(6\theta_{21}^{(\alpha)}) \right). \end{aligned} \quad (2.16)$$

In above equation, $\mathbf{r}$ is the two-dimensional (projected) vector connecting two carbon

atoms, $r = |\mathbf{r}|$ and $\bar{r} = r/a$ where graphene lattice constant $a = 2.46\text{\AA}$. Note that, FK description for inter-layer hopping cannot capture the effect of lattice corrugation. $V_i(r)$ are fitted as:

$$\begin{aligned} V_0(r) &= \lambda_0 e^{-\xi_0 \bar{r}^2} \cos(\kappa_0 \bar{r}), \\ V_3(r) &= \lambda_3 \bar{r}^2 e^{-\xi_3 (\bar{r} - x_3)^2}, \\ V_6(r) &= \lambda_6 e^{-\xi_6 (\bar{r} - x_6)^2} \sin(\kappa_6 \bar{r}), \end{aligned} \quad (2.17)$$

and θ_{12} (θ_{21}) indicates the angle between the projected inter-layer bond $\mathbf{r}$ and nearest neighbour bond of atom-1 (atom-2). α is a bond index. All fitted parameters for FK-TB model is summarized in Table. 2.1 [82, 83, 100, 101].

Based on the theoretical formalism of TAPW, we developed a robust Python package to perform calculation. Numerical schemes including full TB method, TAPW method and BM model are all realized. Their characteristics are briefly summarized in Table.2.2. We plot single particle electronic spectrum of MATBG ($\theta = 1.085^\circ$) using different input structures (rigid, corrugated and relaxed TBG structure) with different methods (TAPW and full TB) and TB parameterization (Slater-Koster (SK) TB model and Fang-Kaxiras (FK) TB model [100]) in Fig. 2.5 for benchmark. TAPW results all present good agreement with the full TB results. Lattice relaxation is performed using molecular dynamics with the force field developed in Appx.A.3.

Table 2.1: Parameters in FK-TB model for TBG.

Intra	t_0 (eV)	α_0	β_0	t_1 (eV)	α_1	r_1
	-18.4295	1.2771	2.3934	-3.7183	6.2194	0.9071
Inter	λ_i (eV)	ξ_i	x_i	κ_i		
V_0	0.3155	1.7543		2.0010		
V_3	-0.0688	-0.0688	0.5212			
V_6	-0.0083	2.8764	1.5206	1.5731		

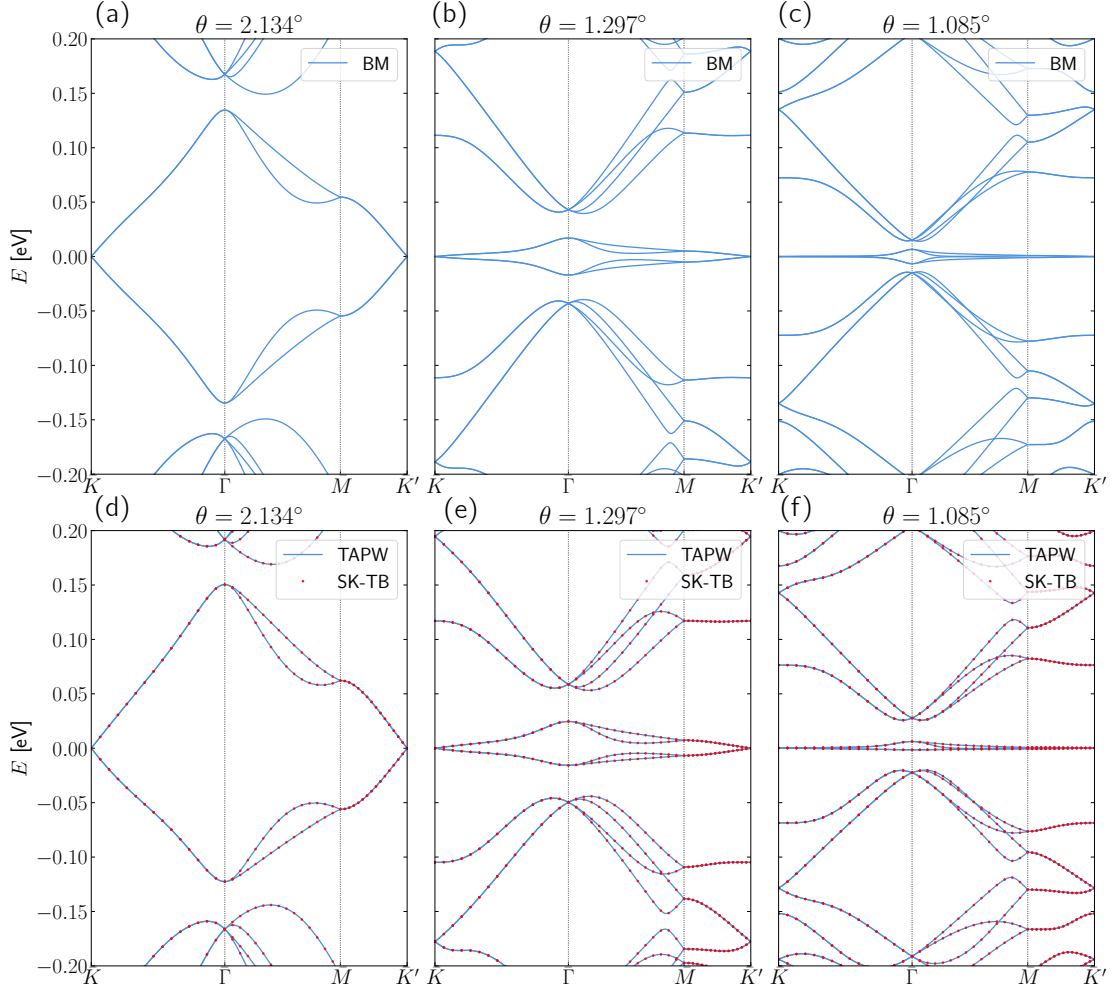


Figure 2.4: Electronic Band structures of TBG along high symmetry points $\bar{K} - \bar{\Gamma} - \bar{M} - \bar{K}'$ at different twist angles using different methods when corrugation effect is taken into consideration. (a)-(c): Band structures computed using BM model for $\theta = 2.134^\circ, 1.297^\circ, 1.085^\circ$, respectively. (d)-(f): Band structures computed using full TB (SK-parameterization) and TAPW method for $\theta = 2.134^\circ, 1.297^\circ, 1.085^\circ$, respectively. The full TB Reference results are denoted in red dots while the TAPW results are in blue lines. TB results show a particle hole asymmetry compared with BM model.

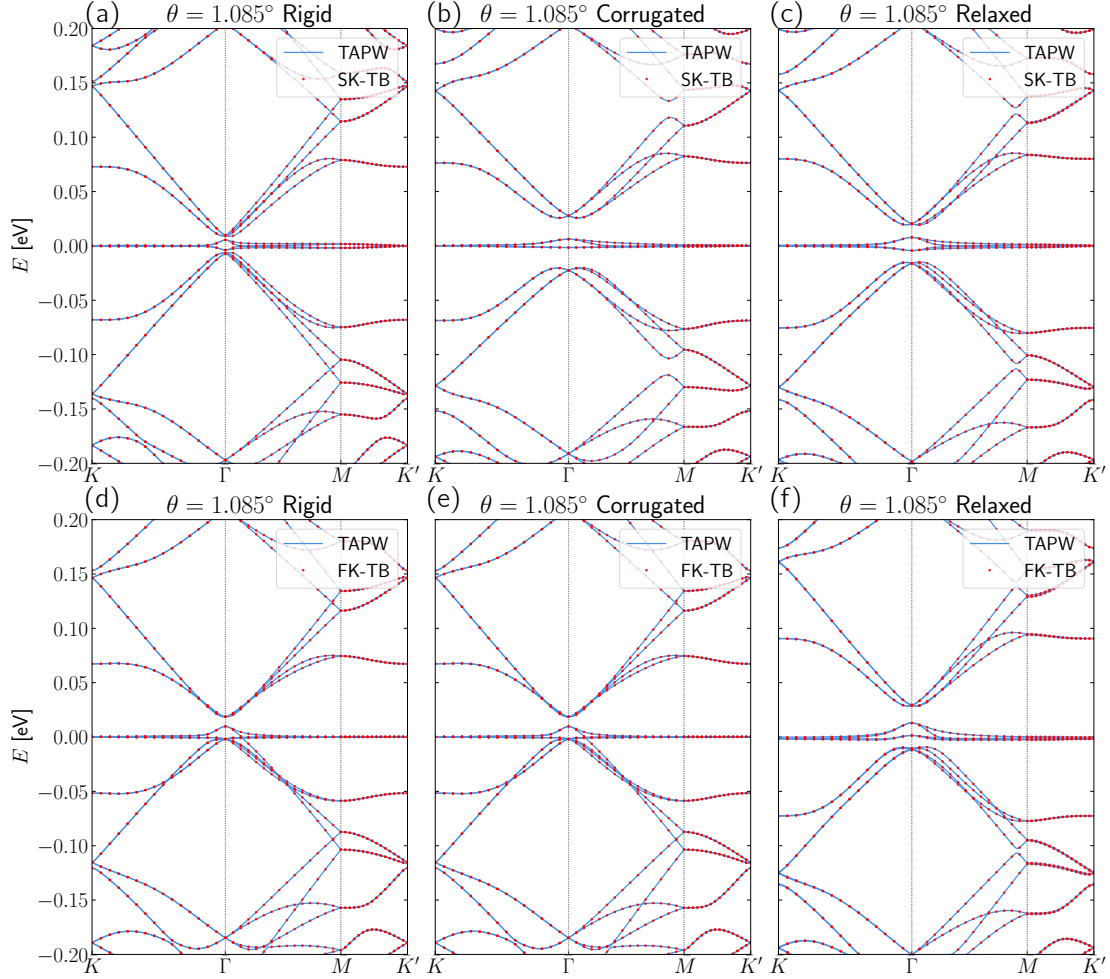


Figure 2.5: Electronic Band structures of MATBG ($\theta = 1.085^\circ$) along high symmetry points $\bar{K} - \bar{\Gamma} - \bar{M} - \bar{K}'$ using different methods and different TB parameters. (a)-(c): Band structures computed using SK-TB model for rigid, corrugated and relaxed structure, respectively. (d)-(f): Band structures computed using FK-TB model for rigid, corrugated and relaxed structure, respectively. The full TB Reference results are denoted in red dots while the TAPW results are in blue lines.

Table 2.2: Comparison between different computational methods in TBG system

Methods	Full TB model	BM Model	TAPW method
Rotation Angle	any	small	small
# of basis	# of atoms	$4 \times$ # of G vectors	$4 \times$ # of G vectors
Cost	High	Low	Medium
Accuracy	High	Medium	High
Corrugation	set coordinates	let $u \neq u'$	set coordinates
Incommensurate System	No	Yes	No
Easy to Use	No	Yes	Yes

We take WannierTools (WT) [25] as a benchmark to present the efficiency of TAPW method. In TB calculation, the whole process can be divided into two parts: setting up the hopping matrix $\mathbf{T}_2$ or hopping integral file `hr_dat` and diagonalizing the TB Hamiltonian. Our numerical strategy takes several seconds to set up the hopping integral matrix $\mathbf{T}_2$ at the first magic angle ($\theta = 1.085^\circ$) while WT takes more than one hour to build the hopping integral file `hr_dat`. TAPW presents a huge advantage when the twist angle gets even smaller, as clarified on the left panel of Fig.2.3. For band structure calculations, we diagonalize the projected matrix instead of struggling with a huge sparse matrix like WT (WT diagonalizes the sparse TB Hamiltonian using Lanczos algorithm integrated in ARPACK [108], this feature is also realized in our Python package.) Our method saves a large amount of time and reproduces low energy band structures perfectly in a wide range of twist angles, as shown in the right panel of Fig.2.3 and Fig.2.4, respectively.

We denote $\mathbf{v}_t$ to be the t -th eigen vector (column vector) of the full TB matrix (t is labeled from Fermi level) and $\bar{\mathbf{v}}_t$ to be the corresponding one of the projected TB matrix. Then we take $\eta = |\mathbf{v}_t^\dagger \cdot \mathbf{X} \cdot \bar{\mathbf{v}}_t|$ as a criteria to evaluate the performance of TAPW basis. The better it will be for TAPW basis if η is closer to 1 ($\mathbf{X} \cdot \bar{\mathbf{v}}$ restores the full

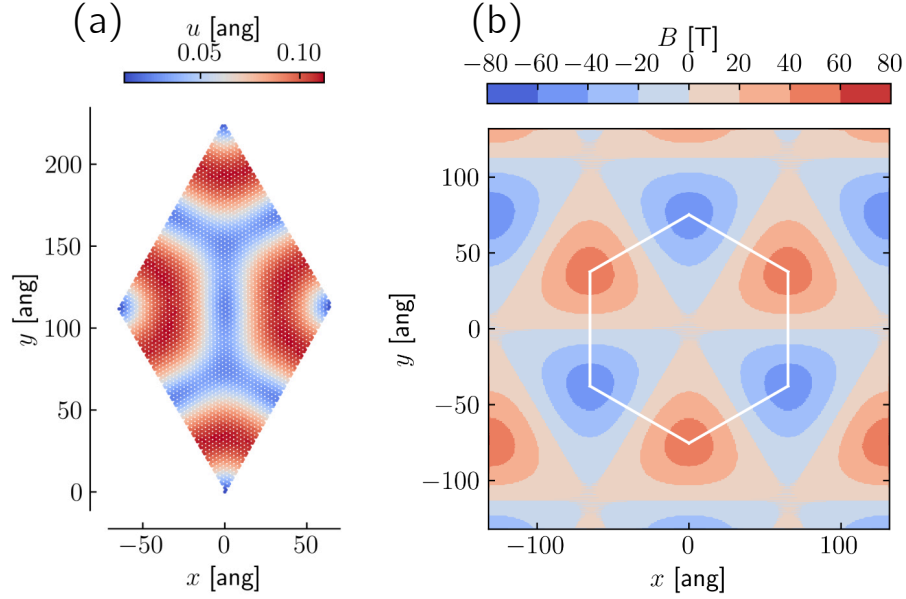


Figure 2.6: (a) In plane lattice relaxation for MATBG depicted in the real space. A vortex pattern appears around the AA stacking region to lower down the elastic energy in the system. (b) Real space distribution of the pseudo magnetic field generated from the lattice relaxation.

tight binding eigenvector). At the first magic angle, we take 244 atomic plane waves per valley (488 bands in total) to expand the full TB matrix and find that $\eta > 99\%$ for eigen vectors of four flat bands at the $\bar{\Gamma}$ point. Now one can be convinced that TAPW method outputs high quality eigen wavefunctions.

2.4 Pseudo magnetic fields and moiré potentials

Compared with BM model, our method generates a similar matrix structure to describe the Hamiltonian if the same G -list is used. It is noteworthy that TAPW method automatically preserves higher order Fourier components of continuum Hamiltonian. In this section, we will show that these higher order terms should be taken into consideration when the twist angle is around 1° , where lattice relaxation plays a vital role.

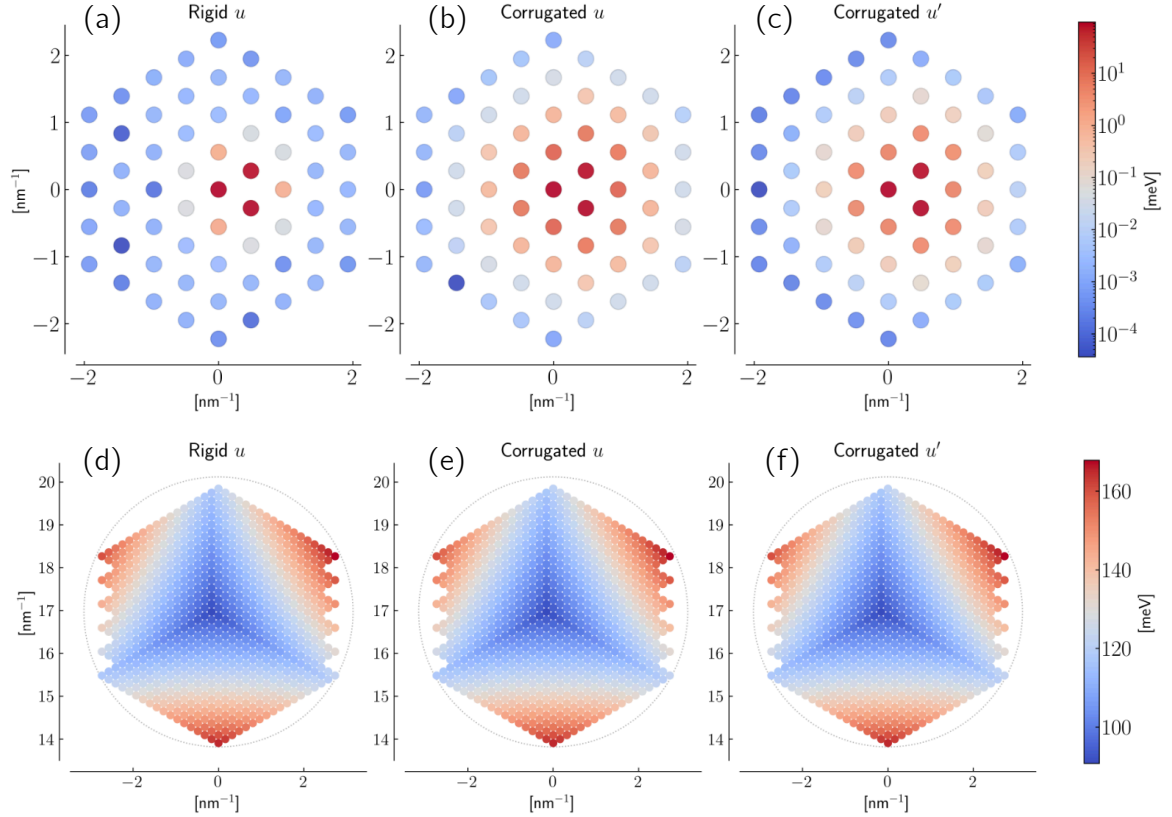


Figure 2.7: Moiré potential $U_{\alpha,\beta}(\mathbf{k}, \mathbf{G})$ calculated using TAPW method when $\theta = 1.085^\circ$. (a)-(c): $\mathbf{G}$ dependence of U when $\mathbf{k}$ is fixed at $\Gamma + \mathbf{G}_0$, $\mathbf{G}_0 = 30(\mathbf{G}_1 + \mathbf{G}_2)$. The largest $\mathbf{G}$ component is located at $\mathbf{G}_0$, $\mathbf{G}_0 - \mathbf{G}_2$, $\mathbf{G}_0 + \mathbf{G}_1$ as the ones from BM. As shown, corrugation effect brings more high order $\mathbf{G}$ components into effect. (d)-(f): $\mathbf{k}$ dependence of U when the largest $\mathbf{G}$ component is reserved which BM totally smears out. (a),(d): Rigid structure as input, $u = |U_{AA}| = |U_{AB}|$. (b),(e): Corrugated structure as input, $u = |U_{AA}|$. (c),(f) Corrugated structure as input, $u' = |U_{AB}|$.

For the intralayer hopping, lattice relaxation is manually taken into consideration in the extended BM model by adding an artificial gauge field $\mathbf{A}(\mathbf{r})$ into the Dirac Hamiltonian $v\boldsymbol{\sigma} \cdot \mathbf{q} \rightarrow v\boldsymbol{\sigma} \cdot (\mathbf{q} + e/\hbar \mathbf{A})$. This gauge field will result in a pseudo magnetic field $B_z = \partial_x A_y - \partial_y A_x$ in the moiré lattice, which can be directly considered in the TAPW formalism. In the case of MATBG, the AA stacking area shrinks and AB stacking area expand when the moiré lattice is fully relaxed, as shown in Fig. 2.6-(a). The carbon atoms moving around the AA stacking area and form a vortex like pattern. The corresponding pseudo magnetic field can be as large as around 80 T in the AB stacking area, as presented in Fig. 2.6-(b). Note that, the pseudo magnetic field cancels out in the whole moiré lattice.

For the interlayer hopping, people do Fourier analysis for $U_{\alpha,\beta}(\mathbf{k}, \mathbf{r})$ in the BM model, as commonly analyzed in Ref. [75]:

$$U_{\alpha,\beta}(\mathbf{k}, \mathbf{r}) = \sum_{m_1, m_2} \tilde{U}_{\alpha,\beta}(m_1 \mathbf{b}_1 + m_2 \mathbf{b}_2 + \mathbf{k}) e^{i(m_1 \mathbf{b}_1 + m_2 \mathbf{b}_2) \cdot \boldsymbol{\delta}_{\alpha,\beta}} e^{i(m_1 \mathbf{G}_1 + m_2 \mathbf{G}_2) \cdot \mathbf{r}}, \quad (2.18)$$

where,

$$\tilde{U}_{\alpha,\beta}(\mathbf{q}) = -\frac{1}{S_0} \int t[\mathbf{R} + d(\mathbf{R} - \boldsymbol{\delta}_{\alpha,\beta})\mathbf{e}_z] e^{-i\mathbf{q} \cdot \mathbf{R}} d\mathbf{R}. \quad (2.19)$$

BM model adopts two important assumptions for the description of moiré potential.

1. $\tilde{U}(\mathbf{q})$ decays in $q \approx 1/r_0$ because the transfer integral t is determined by Slater Koster formula in Eq.(2.14) which exponentially decays in $R \approx r_0$. That's the reason why BM model only conserves three largest Fourier components: $(m_1, m_2) = (0, 0), (-1, 0), (0, 1)$ for $\mathbf{K}$ valley.
2. When performing $\mathbf{k}$ sampling in the area that is close to $\mathbf{K}$, BM model always assumes $U(\mathbf{k}, \mathbf{r}) = U(\mathbf{K}, \mathbf{r})$. It is a good approximation that $\tilde{U}(|\mathbf{k}|) \approx \tilde{U}(|\mathbf{K}|)$ at

the first magic angle when $|\mathbf{k} - \mathbf{K}| \leq |\mathbf{G}| \approx |\mathbf{K}|$ [9], because $|\mathbf{K}| \approx 30|\mathbf{G}| \gg |\mathbf{G}|$.

In common practice, $\mathbf{k}$ sampling is performed in the area that $|\mathbf{k} - \mathbf{K}| \leq 4|\mathbf{G}|$ [75] when calculating the band structure of MATBG. When $|\mathbf{k} - \mathbf{K}| \leq |\mathbf{G}|$, it brings us the accurate flat band picture which is the original BM model [9]. When $|\mathbf{k} - \mathbf{K}| > |\mathbf{G}|$, these assumptions are rough and result in more deviation on the bands away from Fermi surface compared with the full TB results, as shown in Fig.5 of Ref. [98]. Additionally, atomic relaxation at small twist angle will make high order $\mathbf{G}$ components of moiré potential more significant than the rigid one [76, 80, 80–83].

Above analysis suggests a more accurate description for the moiré potential is needed. Different from existing studies which expand BM manually to higher orders [76, 79–83] and integrate a k -linear term in the interlayer coupling to capture the non-local tunneling [79–81], our new method automatically expands the moiré potential into a tensor form $U_{\alpha\mathbf{G}_i,\beta\mathbf{G}_j}(\bar{\mathbf{k}})$, where $\bar{\mathbf{k}} + \mathbf{G}_i = \mathbf{k} \approx \mathbf{K}$. As demonstrated in the first row of Fig.2.7 where we fix $\mathbf{k} = \Gamma + \mathbf{G}_0 \approx 1/2(\mathbf{K}^{(1)} + \mathbf{K}^{(2)})$, corrugation in MATBG brings high order $\mathbf{G}$ components into effect and creates a difference between u and u' . In the second row of Fig.2.7, we see U does rely on $\mathbf{k}$ by conserving the largest $\mathbf{G}$ component of U at different $\mathbf{k}$ points in the area $|\mathbf{k} - \mathbf{K}| < 5|\mathbf{G}|$. These corrections on BM provide us a clear picture for electron and hole asymmetry in the flat bands.

2.5 TAPW method for moiré TMDs

The effective Hamiltonian for moiré transition metal dichalcogenides (TMDs) can be derived by extending the TAPW formalism. Unlike graphene, TMDs lack well-fitted Slater-Koster (SK) formulas due to strong spin-orbit coupling and the involvement of multiple orbitals in bonding. However, an effective first-principles tight-binding Hamil-

tonian can be obtained from ab initio calculations based on numerical atomic orbitals (NAOs). It is noteworthy that modern DFT packages, such as OpenMX and SIESTA, offer an $\mathcal{O}(N)$ algorithm (where N is the number of atoms) using NAO bases, enabling self-consistent DFT calculations.

Once the ab initio tight-binding Hamiltonian is available, we can establish a comprehensive workflow for the first-principles calculation of moiré materials, as illustrated in Fig. 2.9. The initial step involves constructing the full tight-binding matrix via a one-shot DFT calculation. Subsequently, the TAPW transformation can be performed to accelerate the process of predicting the material's topological features and transport properties. Following the TAPW method, a crucial step is constructing the plane wave projection matrix $\mathbf{X}$.

For example, in the case of twisted MoTe_2 , the projected Hamiltonian will have a size of $n_{\mathbf{G}} \times (2 \times n_{\text{Mo-NAO}} + 4 \times n_{\text{Te-NAO}})$ without spin-orbit coupling. This matrix size will double if spin-orbit coupling is included. In the case of rigid 9.43° , we use 19 NAOs for Te and 14 NAOs for Mo, and the DFT band structure can be well converged. As shown in Fig. 2.8, the TAPW band structures show good consistency with the DFT results with only 7 plane waves per valley. Additionally, the bands from the Γ valley are also close to the Fermi energy compared with the case of graphene.

2.6 Conclusion

As a summary, we present TAPW, a carefully optimized numerical scheme to down-fold the full “tight binding” Hamiltonian for TBG to a low energy effective model using a series of truncated atomic plane waves. Our method shows a perfect consistency with the low energy bands solved from full tight binding Hamiltonian, not only for moiré

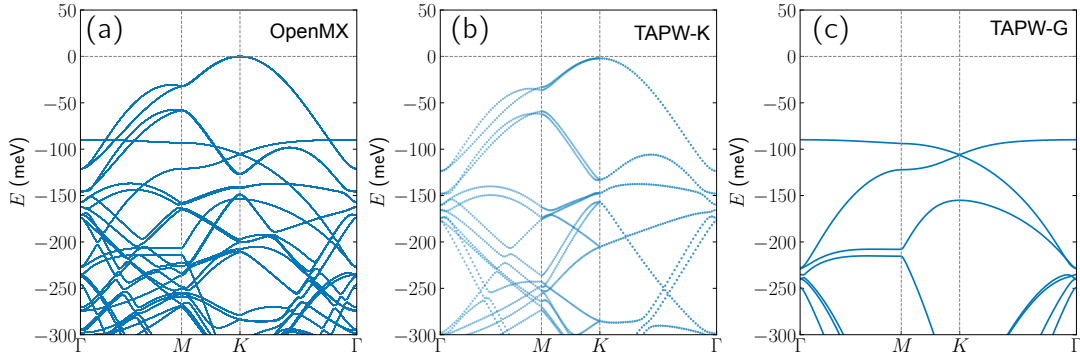


Figure 2.8: Band structures for rigid 9.43° twisted MoTe₂ without SOC. (a) *ab initio* band structure calculation based on self-consistent DFT calculation using OpenMX. (b) projected TAPW band structure using truncated plane waves from two K valleys. (c) projected TAPW band structure using truncated plane waves from Γ valley.

electrons. This kind of low cost projection can be generalized to other twisted moiré systems if credible tight binding Hamiltonian is constructed to describe electrons or faithful dynamic matrix for phonons. For example, parameterized tight binding Hamiltonian for twisted transition metal dichalcogenides (TMDCs) [109, 110] has been proposed recently. Lattice dynamics of several important 2D materials can be simulated using semi-classical molecular dynamics integrated with modern force fields [111, 112]. In this chapter, we further visualize moiré potential of MATBG by projecting interlayer interaction U to a series of atomic plane waves which clarifies the importance of high order G components and related k dependence in the band structure at small twist angles.

TAPW provides a systematical way to study moiré electrons and phonons in a single particle manner. By freezing lattice vibrations induced by specific phonon mode, TAPW can build low energy effective model containing electron phonon coupling [113, 114]. As pointed out in Ref. [115–117], the iTO phonon-induced local Kekulé distortion may correspond to fruitful phase diagrams in MATBG and this kind of distortion has been observed in a recent nano Raman experiment [118]. Another feature of TAPW is the

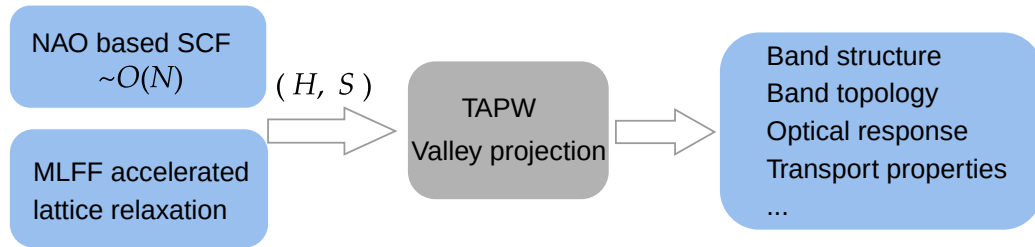


Figure 2.9: The whole workflow of the first principle calculation of moiré materials. The TAPW method can be integrated with first principle calculations and helps accelerate the process of materials discovery and prediction.

model itself is intrinsically equivalent to the generalized BM model. The projected Hamiltonian shares the same structure which makes further computation easy to implement.

Chapter 3

Moiré phonons and electron phonon coupling in the MATBG system

The mathematical structure of the dynamic matrix, which contains all information about phonons, shares many similarities with the tight-binding Hamiltonian describing electrons. We extend the Truncated Atomic Plane Wave formalism discussed earlier to the system of low-frequency moiré phonons and explore their vibrational features in magic-angle twisted bilayer graphene (MATBG). We further highlight that the in-plane transverse (iTO) mode can couple strongly with the flat bands of MATBG. The maximally coupled moiré iTO phonons are constructed by integrating all iTO branches coupling with f electrons of the flat bands.

Some of the contents in this chapter have substantially appeared in Ref. [119], which is in submission.

3.1 Introduction

Magic angle twisted bilayer graphene (MATBG) hosts eight flat bands near the charge neutrality point (CNP) and exhibits captivating properties in transport and optical experiments [119–127]. Numerous studies [128–142] have been dedicated to understanding the ground state and associated symmetry-breaking mechanisms in MATBG’s exotic quantum phases. These investigations typically begin with the original BM model [143], focusing exclusively on local interlayer tunneling. However, lattice reconstruction and uniform strain effects are common in TBG samples [53, 122, 123, 144–148] making non-local interlayer tunneling noteworthy and introducing novel competing orders [149–153]. It is also demonstrated in a new high resolution STM experiment [154] that the incommensurate Kekulé spiral (IKS) [149] is the most probable candidate for the correlated insulator states at filling $\nu = \pm 2$ in typical TBG samples with moderate strain. However, for ultra-low strain samples, the time reversal symmetric intervalley coherent (TIVC) state [152, 155–157], which does not exhibit IKS order and *retains the original moiré periodicity*, is more favorable at $\nu = -2$.

Deviations from a rigid TBG structure at the moiré scale can not only arise from strain effects but also from the system’s inherent vibrational modes, an aspect that has been largely neglected until recently. Two recent experimental observations, one using the nano-Raman technique [127] and the other using μ -ARPES [119], have provided theorists with some hints that optical phonons may play important roles in the MATBG system. In the nano-Raman experiment, two strong optical signals appear at around 1580 cm^{-1} (195 meV) and 2650 cm^{-1} ($2 \times 165 \text{ meV}$), and the phonon-induced vibrations are well-localized at the AA-stacking area. These signals correspond to an in-plane longitudinal optical (iLO) phonon process (G band) and a two-in-plane transverse optical (iTTO) phonon process ($2G$ band) [158]. In the μ -ARPES experiment, phonon-

induced replica bands with equal energy spacing (150 ± 15 meV) are observed in superconducting MATBG samples unaligned with hBN substrate, while no such phenomena appear in non-superconducting MATBG samples with hBN substrates. Theoretically, the importance of electron-phonon coupling in the MATBG system has been revealed in a series of works [159–165] to understand the unconventional superconductivity. Based on large-scale molecule dynamics simulations and microscopic tight-binding calculations [53, 160, 163, 165, 166], the density of states (DOS) of the optical phonon spectrum for MATBG is almost the same as that of Bernal bilayer graphene, suggesting that the moiré potential can be safely ignored for high energy phonon modes. The Eliashberg function, which indicates the strength of electron-phonon coupling, has two peaks originating from optical phonons located at the Γ point and the K point of monolayer graphene [160, 165]. However, there is still no detailed description for how these optical moiré phonons influence the lattice dynamics and couple with the low energy electrons due to the complexity of the moiré system.

Building on these experimental measurements, we will conduct a comprehensive investigation of electron-phonon interactions within the MATBG system. Our analysis begins with a continuous electron-phonon coupling (EPC) description, examining how these optical phonons interact with the zeroth pseudo-Landau levels (ZLLs) of MATBG [167, 168] based on symmetry considerations. We find that A_1/B_1 mode from graphene’s K valley effectively correlate with localized low-energy electrons. The i TO A_1/B_1 mode, initially identified in Ref. [166], introduces a Kekulé-like distortion in real space. Moreover, the EPC strength can be substantially enhanced [169–172] in the moiré system due to high-order Umklapp processes. By performing frozen phonon calculations with atomistic tight-binding models for different orders of Umklapp processes, we identify their decaying characteristics, which are entirely overlooked in

Refs. [153, 166, 173]. We further construct the maximally coupled Wannier moiré phonons, which exhibit a well-localized feature at the moiré scale, by integrating all effective EPC. This finding aligns with the nano-Raman experiment and STM experiment, which have shown strong signals at the AA-stacking region of the TBG.

3.2 Phonons and its “tight binding” description

3.2.1 Equation of motion for lattice vibrations

We will first briefly discuss lattice vibrations in the language of classical mechanics. Let’s define a general lattice structure, for atom $i\alpha$ (here we use a joint index $i\alpha$ to describe atom positions which makes it easy to describe carbon atoms in Twisted Bilayer Graphene system, α is an index for sublattice A_1, A_2, B_1, B_2 , i is an index for the graphene lattice in the moiré unit cell), in lattice I, the momentary position of the atom $i\alpha$ should be,

$$\mathbf{x}_{Ii\alpha}(t) = \mathbf{R}_I + \boldsymbol{\tau}_{i\alpha} + \mathbf{u}(\mathbf{R}_{Ii\alpha})(t), \quad (3.1)$$

with $\boldsymbol{\tau}_{i\alpha}$ being sublattice positions and $\mathbf{u}_{i\alpha}$ being displacement induced by lattice vibrations. The classical Hamiltonian for describing lattice vibrations under harmonic approximation is,

$$H = \frac{1}{2M_{i\alpha}} \sum_{I\alpha\mu} p_\mu(\mathbf{R}_{Ii\alpha}) p_\mu(\mathbf{R}_{Ii\alpha}) + \frac{1}{2} \sum_{I\alpha\mu} \sum_{Jj\beta\nu} \Phi_{\mu\nu}(\mathbf{R}_{Ii\alpha} - \mathbf{R}_{Jj\beta}) u_\mu(\mathbf{R}_{Ii\alpha}) u_\nu(\mathbf{R}_{Jj\beta}). \quad (3.2)$$

Here, we define $\mathbf{R}_{Ii\alpha} = \mathbf{R}_I + \boldsymbol{\tau}_{i\alpha}$ for short and $M_{i\alpha}$ is the mass for atom- α . Φ is a 3×3 force constant describing the effective mechanical interaction between two atoms. The Hamiltonian is quadratic, so it must be equivalent to a set of harmonic oscillators. By

using Hamilton's equation, the equation of motion now reads,

$$M_{i\alpha} \frac{d^2 u_\mu(\mathbf{R}_{i\alpha})}{dt^2} = - \sum_{\nu Jj\beta} \Phi_{\mu\nu}(\mathbf{R}_{i\alpha} - \mathbf{R}_{Jj\beta}) u_\nu(\mathbf{R}_{Jj\beta}). \quad (3.3)$$

One can further write the equation of motion for the phonon field $\mathbf{u}$ in the frequency domain after performing Fourier Transformation,

$$\frac{1}{M_{i\alpha}} \sum_{Jj\beta\nu} \Phi_{\mu\nu}(\mathbf{R}_{i\alpha} - \mathbf{R}_{Jj\beta}) u_\nu(\mathbf{R}_{Jj\beta}) = \omega^2 u_\mu(\mathbf{R}_{i\alpha}). \quad (3.4)$$

To bring Eq. (3.4) to a standard form of eigen-value problem, then let $u_\mu(\mathbf{R}_{i\alpha}) = \varepsilon_\mu(\mathbf{R}_{i\alpha}) / \sqrt{M_{i\alpha}}$, one obtains,

$$\sum_{Jj\beta\nu} \frac{1}{\sqrt{M_{i\alpha} M_{j\beta}}} \Phi_{\mu\nu}(\mathbf{R}_{i\alpha} - \mathbf{R}_{Jj\beta}) \varepsilon_\nu(\mathbf{R}_{Jj\beta}) = \omega^2 \varepsilon_\mu(\mathbf{R}_{i\alpha}). \quad (3.5)$$

If we sum over $\varepsilon_\mu^*(\mathbf{R}_{i\alpha})$ on both sides and denote $\hat{\Phi}_{\mu\nu}(\mathbf{R}_{i\alpha} - \mathbf{R}_{Jj\beta}) = \Phi_{\mu\nu}(\mathbf{R}_{i\alpha} - \mathbf{R}_{Jj\beta}) / \sqrt{M_{i\alpha} M_{j\beta}}$,

$$\sum_{i\alpha\mu} \sum_{Jj\beta\nu} \varepsilon_\mu^*(\mathbf{R}_{i\alpha}) \hat{\Phi}_{\mu\nu}(\mathbf{R}_{i\alpha} - \mathbf{R}_{Jj\beta}) \varepsilon_\nu(\mathbf{R}_{Jj\beta}) = \omega^2. \quad (3.6)$$

The above equation is a simple eigen-value problem, however, so many harmonic oscillators are coupled together which should be simplified using translational symmetry of the solid state system. By taking advantage of the periodic boundary condition of the supercell and the Bloch theorem, one can get,

$$\begin{aligned} \varepsilon_\mu(\mathbf{R}_{i\alpha}) &= \frac{1}{\sqrt{N}} \sum_{\bar{\mathbf{q}} \in \text{B.Z.}} e^{i\bar{\mathbf{q}} \cdot \mathbf{R}_{i\alpha}} \tilde{\varepsilon}_{i\alpha\mu}(\bar{\mathbf{q}}) \\ \sum_{\bar{\mathbf{q}}} \tilde{\varepsilon}^T(\bar{\mathbf{q}}) \cdot \mathbf{D}(\bar{\mathbf{q}}) \cdot \tilde{\varepsilon}(\mathbf{q}) &= \omega^2. \end{aligned} \quad (3.7)$$

The entries of the dynamic matrix depend on the chosen gauge convention. In the atomic gauge convention (as the one used in PhononPy), the entries of $\mathbf{D}$ are defined as follows,

$$D_{i\alpha\nu,j\beta\mu}(\bar{\mathbf{q}}) = \sum_{\mathbf{J}} \hat{\Phi}_{\mu\nu}(\mathbf{0} + \boldsymbol{\tau}_{i\alpha} - \mathbf{R}_{\mathbf{J}j\beta}) e^{i\bar{\mathbf{q}} \cdot (\mathbf{R}_{\mathbf{J}} + \boldsymbol{\tau}_{j\beta} - \boldsymbol{\tau}_{i\alpha})}. \quad (3.8)$$

The characteristic phonon mode (polarization phonon mode) $\boldsymbol{\varepsilon}_{i\alpha}(\bar{\mathbf{q}}, b)$ together with characteristic vibration frequency ω_b can be retrieved by solving the eigenvalue problem. Then the general vibration $\mathbf{u}(\mathbf{R}_{i\alpha})(t)$ of the system can be expanded as:

$$\mathbf{u}(\mathbf{R}_{i\alpha})(t) = \frac{1}{\sqrt{NM_{i\alpha}}} \sum_{\bar{\mathbf{q}}, b} Q_{\bar{\mathbf{q}}, b} \boldsymbol{\varepsilon}_{i\alpha}(\bar{\mathbf{q}}, b) e^{i\bar{\mathbf{q}} \cdot \mathbf{R}_{i\alpha}} e^{i\omega_b(\bar{\mathbf{q}})t}, \quad (3.9)$$

where $Q_{\bar{\mathbf{q}}, b}$ is the co-called normal coordinates. Phonons and electrons in solids share several similarities in their descriptions, although there are fundamental differences between them. Phonons, as collective vibrational modes of the crystal lattice, are bosons, while electrons are fermions. In the context of solid-state systems, the force constant matrix for phonons plays a role similar to the hopping matrix in the tight-binding formalism for electrons. The force constant matrix describes the interatomic interactions and governs the dynamics of lattice vibrations, just as the hopping matrix characterizes the electron hopping between neighboring atomic sites in the tight-binding model.

To establish a connection between the tight-binding description for electrons and phonons, it is essential to comprehend the concept of the “Wannier basis” in the context of phonons. The hopping matrix element in electronic problem $\langle \mathbf{R}_{\mathbf{I}} + \boldsymbol{\tau}_{i\alpha} | H | \mathbf{R}_{\mathbf{J}} + \boldsymbol{\tau}_{j\beta} \rangle$ is written in the Wannier basis and the reduced force constant $\hat{\Phi}_{\mu\nu}(\mathbf{R}_{i\alpha} - \mathbf{R}_{j\beta})$ is constructed under natural basis $(\mu, \nu = x, y, z)$ of each atomic site. We denote this kind of natural basis as $|\mathbf{R}_{\mathbf{I}} + \boldsymbol{\tau}_{i\alpha\mu}\rangle$. The tight binding basis for the moiré phonons is

constructed using Bloch sum as the one for electrons,

$$|\psi_{i\alpha\mu}(\bar{\mathbf{q}})\rangle = \frac{1}{\sqrt{N_m}} \sum_{\mathbf{R}_I} e^{i\bar{\mathbf{q}} \cdot (\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha})} |\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha}\rangle. \quad (3.10)$$

The phononic “Hamiltonian” can be expanded as:

$$D_{i\alpha\mu,j\beta\nu}(\bar{\mathbf{q}}) = \sum_{\mathbf{R}} e^{-i\bar{\mathbf{q}} \cdot (\boldsymbol{\tau}_{i\alpha} - \boldsymbol{\tau}_{j\beta} - \mathbf{R})} \hat{\Phi}_{\mu\nu}(\boldsymbol{\tau}_{i\alpha} - \boldsymbol{\tau}_{j\beta} - \mathbf{R}) \quad (3.11)$$

and this is the so called dynamic matrix. Now, we have built the one to one map between electronic tight binding description and phononic tight binding description.

3.2.2 A minimal model for phonons in Graphene

Monolayer graphene possesses a honeycomb lattice structure consisting of two inequivalent carbon atoms labeled as A and B. The hexagonal symmetry of the lattice belongs to the point group D_6 . To investigate the lattice dynamics in graphene systems, a theoretical scheme described in Section 3.2.1 can be employed.

In this scheme, the lattice dynamics problem can be tackled by constructing the dynamic matrix D . There are several approaches to obtain the dynamic matrix, such as density functional perturbation theory (DFPT) or molecular dynamics (MD) simulations. However, in this case, we will start with a minimal model where all atoms are connected by springs. Despite its simplicity, this model can capture the basic physics of graphene.

By considering the interactions between adjacent atoms as springs, we can first construct the force constant matrix Φ for the system. The specific values of the force constants can be determined based on empirical knowledge. For a 2D hexagonal lattice,

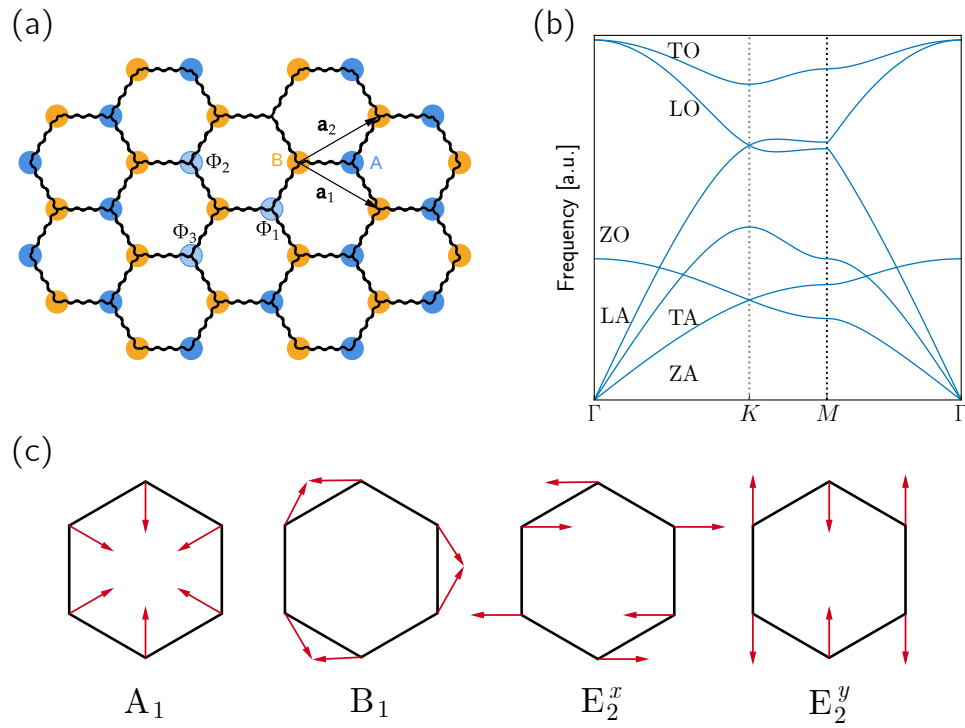


Figure 3.1: (a) Lattice geometry of the monolayer Graphene. Two carbon atoms in the same unit cell are labeled in different colors. (b) Phonon bands for graphene calculated by a simple spring model with only nearest neighbour force constant. The shape of the bands agree well with the ones calculated using molecular dynamics. The names for different phonon branches are also denoted. (c) The vibrational mode for iTO phonon at $\mathbf{K}$ points (A_1/B_1) and degenerate iTO/iLO phonons at Γ point E_{2x}/E_{2y} .

we can calculate the force constant for the system based on the assumption that the atomic potential between two atoms only depends on their relative distance $|\mathbf{r}_1 - \mathbf{r}_2|$ and is given by $V(|\mathbf{r}_1 - \mathbf{r}_2|) = \frac{1}{2}k|\mathbf{r}_1 - \mathbf{r}_2|^2$, which corresponds to a simple spring model. Then the corresponding matrix entries for Φ can be derived as,

$$\begin{aligned}\Phi_{xx}^1 &= \partial_{x_1} \partial_{x_2} V|_{(0,0,0),(a,0,0)} = -V'', \\ \Phi_{yy}^1 &= \partial_{y_1} \partial_{y_2} V|_{(0,0,0),(a,0,0)} = -\frac{V'}{a}, \\ \Phi_{zz}^1 &= \partial_{z_1} \partial_{z_2} V|_{(0,0,0),(a,0,0)} = -\frac{V'}{a}.\end{aligned}\tag{3.12}$$

In the above calculation, we utilized

$$\partial_{x_1} \partial_{x_2} V = -V'' \frac{(x_1 - x_2)^2}{|\mathbf{r}_1 - \mathbf{r}_2|^2} - V' \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} + V' \frac{(x_1 - x_2)^2}{|\mathbf{r}_1 - \mathbf{r}_2|^3},\tag{3.13}$$

and the off-diagonal part of the force constant is zero. If we only consider the nearest neighbours:

$$\begin{aligned}\Phi^2 &= R \left(\theta = \frac{2\pi}{3} \right) \Phi^1 R^{-1} \left(\theta = \frac{2\pi}{3} \right) \\ \Phi^3 &= R \left(\theta = \frac{4\pi}{3} \right) \Phi^1 R^{-1} \left(\theta = \frac{4\pi}{3} \right)\end{aligned}\tag{3.14}$$

Φ_0 can be determined using acoustic sum rule $\Phi_0 + \Phi_1 + \Phi_2 + \Phi_3 = 0$.

Phonon band structure for graphene along high symmetry path is plotted in Fig.3.1. Two inequivalent carbon atoms moving in the three dimensional space brings six degrees of freedom which generates six phonon branches. Three of these branches are acoustic (A) and the left are optical (O). For one acoustic branch and one optical branch, the atomic vibrations are perpendicular to the graphene plane which correspond to out of plane phonon modes (o). For two acoustic branches and two optical

branches, the atomic vibrations are inplane (i). The phonon eigenvectors which also shares a Bloch wave form, are labeled with atomic displacement $\tilde{u}$ multiplied by a phase factor $e^{i\mathbf{q}\cdot\mathbf{r}}$. The inplane atomic displacements (i) can be longitudinal (L) or transversal (T) with respect to direction of $\mathbf{q}$ while the out-of-plane ones should be transversal phonons (T). Then, these six phonon branches are named as oTA, iTA, iLA, oTO, iTO, iLO.

Raman spectroscopy serves as a crucial experimental technique for investigating the vibrational properties of graphene-based systems. In the phonon band structure of monolayer graphene, the iLO (in-plane longitudinal optical) branch intersects with the iTO (in-plane transverse optical) branch at the Brillouin zone center, corresponding to the E_{2g} representation of the D_{6h} point group, which is Raman active. The iTO branch in the vicinity of the Dirac $\mathbf{K}$ point, belonging to the A_1 representation of the D_{3h} point group, also plays a significant role in Raman spectroscopy.

The prominent Raman spectral features for monolayer graphene include the G band, located around 1580 cm^{-1} , and the G' band, situated near 2700 cm^{-1} , using a laser excitation energy of 2.4 eV . The G band arises from the degenerate phonon modes (iLO and iTO) at the Γ point, resulting from first-order Raman scattering processes. The G' band is associated with iTO phonons in the vicinity of the $\mathbf{K}/\mathbf{K}'$ points, involving a second-order cross-valley phonon scattering process (involving either two iTO phonons or one iTO phonon scattered with a defect). Additionally, one may observe a defect-induced D band at approximately half the frequency of the G' band (around 1350 cm^{-1}). Moiré phonons [174–177] also received great attention as novel collective phenomena are observed in twisted bilayer graphene [118] and twisted MoS_2 [178, 179] using Raman spectra technique.

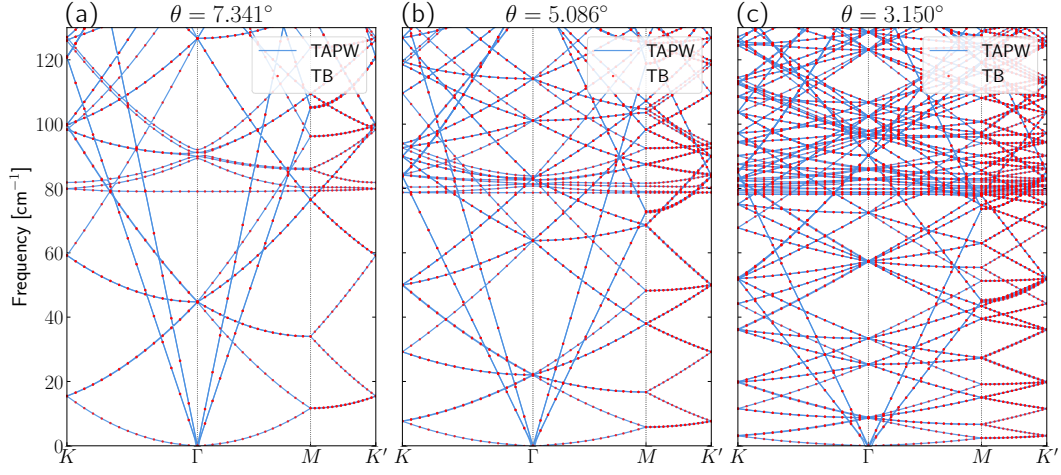


Figure 3.2: Low frequency phonon band structures of TBG along high symmetry points $\bar{K} - \bar{\Gamma} - \bar{M} - \bar{K}'$ at different twist angles, (a): $\theta = 7.341^\circ$, (b): $\theta = 5.086^\circ$, (c): $\theta = 3.150^\circ$. The full tight binding results are denoted in red dots while TAPW bands are in blue lines. The dispersionless layer breathing (LB) mode is located at $\omega \approx 80 \text{ cm}^{-1}$.

3.3 Low frequency moiré phonons

We start to formalize the description the low energy moiré phonons. It is natural to generalize our TAPW method for moiré phonons by mapping tight binding Hamiltonian for electrons to dynamic matrix for lattice vibrations. In this section, we provide a rigorous derivation of TAPW method for moiré phonons and study the low frequency moiré phonons for TBG systems as an example.

The equation of motion for the phonon field $u_\nu(\mathbf{R}_{Jj\beta})$ for TBG under harmonic approximation can be written as:

$$\frac{1}{M_c} \sum_{Jj\beta\nu} \Phi_{\mu\nu}(\mathbf{R}_{Ii\alpha} - \mathbf{R}_{Jj\beta}) u_\nu(\mathbf{R}_{Jj\beta}) = \omega^2 u_\mu(\mathbf{R}_{Ii\alpha}). \quad (3.15)$$

M_c is the mass for the carbon atoms and $\Phi_{\mu\nu}(\mathbf{R}_{Ii\alpha} - \mathbf{R}_{Jj\beta})$ is the force constant between two carbon atoms and μ, ν represent for the Cartesian coordinates. After Fourier

Transforming the phonon field taking advantage of the moiré periodicity and Born–von Karman boundary condition, we get,

$$u_\mu(\mathbf{R}_{I\alpha}) = \frac{1}{\sqrt{N_m}} \sum_{\bar{\mathbf{q}} \in \text{m.B.Z.}} \tilde{u}_{i\alpha\mu}(\bar{\mathbf{q}}) e^{i\bar{\mathbf{q}} \cdot \mathbf{R}_{I\alpha}}, \quad (3.16)$$

which leads to the full tight binding description for moiré phonons,

$$\sum_{j\beta\nu} D_{i\alpha\mu,j\beta\nu}(\bar{\mathbf{q}}) \tilde{u}_{j\beta\nu}(\bar{\mathbf{q}}) = \omega^2(\bar{\mathbf{q}}) \tilde{u}_{i\alpha\mu}(\bar{\mathbf{q}}), \quad (3.17)$$

and the dynamic matrix $D_{i\alpha\mu,j\beta\nu}(\bar{\mathbf{q}})$ defined in the moiré B.Z. is,

$$D_{i\alpha\mu,j\beta\nu}(\bar{\mathbf{q}}) = \frac{1}{M_c} \sum_{\mathbf{R}_J} \Phi_{\mu\nu}(\mathbf{0} + \boldsymbol{\tau}_{i\alpha} - \mathbf{R}_{Jj\beta}) e^{i\bar{\mathbf{q}} \cdot (\mathbf{R}_J + \boldsymbol{\tau}_{j\beta} - \boldsymbol{\tau}_{i\alpha})}. \quad (3.18)$$

One should keep in mind that in moiré systems, there are two kinds of periodicity if the inter-layer coupling is turned off. The first one is the moiré periodicity and the second one is the atomic periodicity. For moiré systems like TBG, we can always expand phonon fields using the periodicity of single layer graphene,

$$\begin{aligned} u_\mu(\mathbf{R}_{I\alpha}) &= \frac{1}{\sqrt{N}} \sum_{\mathbf{q} \in \text{B.Z.}} \bar{u}_{\alpha\mu}(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{R}_{I\alpha}} \\ &= \frac{1}{\sqrt{N_m N_a}} \sum_{\bar{\mathbf{q}}, \mathbf{G}_n} \bar{u}_{\alpha\mu}(\bar{\mathbf{q}} + \mathbf{G}_n) e^{i(\bar{\mathbf{q}} \cdot \mathbf{R}_{I\alpha} + \mathbf{G}_n \cdot \boldsymbol{\tau}_{i\alpha})}. \end{aligned} \quad (3.19)$$

Substitute Eq. (3.19) to the equation of motion for the phonon field and use the definition of dynamic matrix $D_{i\alpha\mu,j\beta\nu}(\bar{\mathbf{q}})$, we get the following equation,

$$\sum_{\beta\nu m} \bar{D}_{\alpha\mu n, \beta\nu m}(\bar{\mathbf{q}}) \bar{u}_{\beta\nu}(\bar{\mathbf{q}} + \mathbf{G}_m) = \omega^2(\bar{\mathbf{q}}) \bar{u}_{\alpha\mu}(\bar{\mathbf{q}} + \mathbf{G}_n), \quad (3.20)$$

where the projected dynamic matrix $\bar{D}_{\alpha\mu n, \beta\nu m}(\bar{\mathbf{q}})$ is defined as,

$$\bar{D}_{\alpha\mu n, \beta\nu m}(\bar{\mathbf{q}}) = \frac{1}{N_a} \sum_{ij} e^{-i\mathbf{G}_n \cdot \boldsymbol{\tau}_{i\alpha}} D_{i\alpha\mu, j\beta\nu}(\bar{\mathbf{q}}) e^{i\mathbf{G}_m \cdot \boldsymbol{\tau}_{j\beta}}. \quad (3.21)$$

The corresponding matrix form reads:

$$\begin{aligned} \bar{\mathbf{D}}^{\text{TAPW}} &= \sum_{\alpha\beta} \mathbf{X}_\alpha^\dagger \mathbf{D}_{\alpha\beta} \mathbf{X}_\beta \\ &= \mathbf{X}^\dagger \mathbf{D} \mathbf{X}, \end{aligned} \quad (3.22)$$

and α, β now are joint indices for Cartesian coordinates index, A/B sublattice index and layer index. The TAPW formalism for moiré phonons shares a lot of similarities with the one describing electrons. One the force constant which is the hopping matrix in the case of electron, is accurately calculated, the phonons can be easily solved. For low frequency phonon bands, we construct a truncated $\mathbf{G}$ -list centered at Γ point instead of $\mathbf{K}$ or $\mathbf{K}'$ point in the problem of electrons.

We use frozen phonon method to compute the force constants $\Phi_{\mu\nu}(\mathbf{R}_{Ii\alpha} - \mathbf{R}_{Jj\beta})$ from the relaxed structure:

$$\Phi_{\mu\nu}(\mathbf{R}_{Ii\alpha} - \mathbf{R}_{Jj\beta}) = \frac{\partial^2 U}{\partial \mathbf{R}_{\mu Ii\alpha} \partial \mathbf{R}_{\nu Jj\beta}} = -\frac{\partial \mathbf{F}_{\nu Jj\beta}}{\partial \mathbf{R}_{\mu Ii\alpha}}. \quad (3.23)$$

where $U = U_{\text{bonded}} + U_{\text{non-bonded}}$ is the potential energy consisting of the bonded intra-layer interactions and non-bonded van der Waals inter-layer interactions. The bonded interactions can be modelled by the Dreiding force fields [180], and the non-bonded van der Waals interactions are modelled by an exponential-6 form [180, 181]. Details are described in Appx.A.3. Before computing the force constants, the lattice was relaxed to optimize the geometry by performing conjugate gradient (CG) algorithm em-

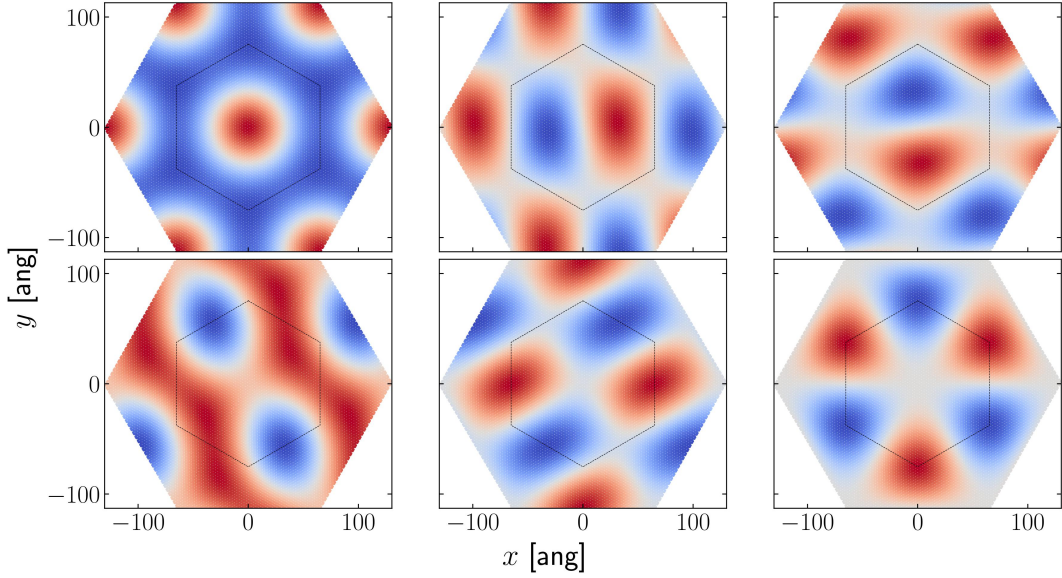


Figure 3.3: Low frequency optical phonon modes solved at $\bar{\Gamma}$ point of MATBG using TAPW method. The black hexagon marks the Wigner–Seitz cell of TBG. We find all-symmetric-type, dipolar-type, quadrupolar-type and octupolar-type out-of-plane vibrations as in Ref. [3].

bedded in LAMMPS [182]. With relaxed lattice, the force constants can be approximated by finite displacement method [183] with a small displacement of 0.01 Å.

The full dynamic matrix $\mathbf{D}$ can then be constructed like full TB Hamiltonian for electrons:

$$\mathbf{D} = \mathbf{T}_1 * \Phi, \quad (3.24)$$

which is an element-wise product with $\mathbf{T}_1$ being hopping phase matrix. As shown in Fig. 3.2, low frequency phonon bands solved at a series of twist angles of TBG using our TAPW method show a perfect consistency with the ones from directly diagonalizing dynamic matrix $\mathbf{D}$. Furthermore, polarization vector $\tilde{u}_{i\alpha\mu}(\bar{\mathbf{q}})$ can be restored using eigen vectors $\bar{u}_{\alpha\mu}(\bar{\mathbf{q}} + \mathbf{G}_n)$ of $\bar{\mathbf{D}}^{\text{TAPW}}$:

$$\tilde{u}_{i\alpha\mu}(\bar{\mathbf{q}}) = \frac{1}{\sqrt{N_a}} \sum_n \bar{u}_{\alpha\mu}(\bar{\mathbf{q}} + \mathbf{G}_n) e^{i\mathbf{G}_n \cdot \boldsymbol{\tau}_{i\alpha}}, \quad (3.25)$$

which has a simple matrix form $\tilde{\mathbf{u}} = \mathbf{X}\bar{\mathbf{u}}$. We plot out-of-plane vibrations resulting from low frequency optical phonon modes of MATBG at $\bar{\Gamma}$ point (around 0-30 cm^{-1}) using our TAPW method in Fig.3.3. The result is consistent with the ones calculated in Ref. [3] where the authors directly solve a huge dynamic matrix (33492×33492). In our calculation, we use 732 atomic plane waves centered at Γ point to expand the full dynamic matrix and the projected dynamic matrix is reduced to 732×732 . Then we can finish the whole computation on a laptop. (The number of atomic plane waves is $61 \times 12 = 732$ when $N_G = 61$.) Based on above numerical experiments, we believe that TAPW method can not only provide accurate band structures but also detailed eigen wavefunctions for moiré phonons.

3.4 Optical moiré phonons

3.4.1 Minimal Hamiltonian with electron phonon coupling

Previous studies [3, 113, 114] based on large scale molecular dynamics (MD) simulations point out that the density of states of MATBG at high frequency is almost the same as bilayer graphene, suggesting that the impact of moiré potential on high-frequency phonons is negligible. Then, we can mimic phonons at moiré Γ_M point, which result from iTO/iLA/iLO phonons at graphene's K/K' points through a folding. For layer-symmetric iTO A_1/B_1 phonons, the distortion field under the $k \cdot p$ theory can

be written down as,

$$\begin{aligned} \mathbf{u}_l^{A1}(\mathbf{Q}) &\sim s \left(\sum_{\{\mathbf{Q}\}} e^{i(\mathbf{K}_l^+ + \mathbf{Q}) \cdot \mathbf{r}} \mathbf{e}_l^+ + \sum_{\{\mathbf{Q}\}} e^{i(\mathbf{K}_l^- + \mathbf{Q}) \cdot \mathbf{r}} \mathbf{e}_l^- \right), \\ \mathbf{u}_l^{B1}(\mathbf{Q}) &\sim is \left(\sum_{\{\mathbf{Q}\}} e^{i(\mathbf{K}_l^+ + \mathbf{Q}) \cdot \mathbf{r}} \mathbf{e}_l^+ - \sum_{\{\mathbf{Q}\}} e^{i(\mathbf{K}_l^- + \mathbf{Q}) \cdot \mathbf{r}} \mathbf{e}_l^- \right), \end{aligned} \quad (3.26)$$

where $\mathbf{e}_{\pm,A} = (\pm i/2, -1/2)$, $\mathbf{e}_{\pm,B} = (\pm i/2, 1/2)$ for A/B sublattices, $\pm$ represents two valleys, this describes the polarization vector of iTO phonon for monolayer graphene located at the $\mathbf{K}$ point and s characterizes the distortion strength. This approximation is valid since the moiré reciprocal lattice is much smaller than the atomic one. Note that the distortion field we construct obeys the Bloch theorem and does not break the original translational symmetry of the pristine cell because $\mathbf{K}_l^\pm + \mathbf{Q}$ is a moiré reciprocal lattice vector. This allows us to perform frozen phonon calculations to estimate the EPC strength using an atomistic tight-binding model. Here, $\{\mathbf{Q}\}$ represents a set of waves vectors that preserve the corresponding irreducible representation (A_1/B_1), see Fig.3.7-(a) for illustration. For the minimal model incorporating phonon mode, we take $\{\mathbf{Q}\}$ to be the nearest ones to the $\mathbf{K}$ point. The EPC strengths can be estimated by feeding updated atomic positions to TAPW Hamiltonian matrix $\mathbf{H}_{\mathbf{k}}^{(\text{TAPW})}$ after considering in-plane lattice distortion, denoted as $\mathbf{H}_{\mathbf{k}}^{\text{eph}}$. By comparing the relative change of phonon-influenced electronic Hamiltonian matrix (at K_M and Γ_M points), $\|\mathbf{H}_{\mathbf{k}}^{\text{eph}} - \mathbf{H}_{\mathbf{k}}^{(\text{TAPW})}\|_2 / \|\mathbf{H}_{\mathbf{k}}^{(\text{TAPW})}\|_2$ (where $\|\cdot\|_2$ represents for L_2 norm of the matrix), we find the flat-band electrons have much stronger coupling (reflected by the slop of energy over displacement) with iTO moiré phonons than other ones (iLO/iLA), as shown in panel (b) of Fig.3.4.

Based on the frozen-phonon estimations, we restrict ourselves to the EPC induced

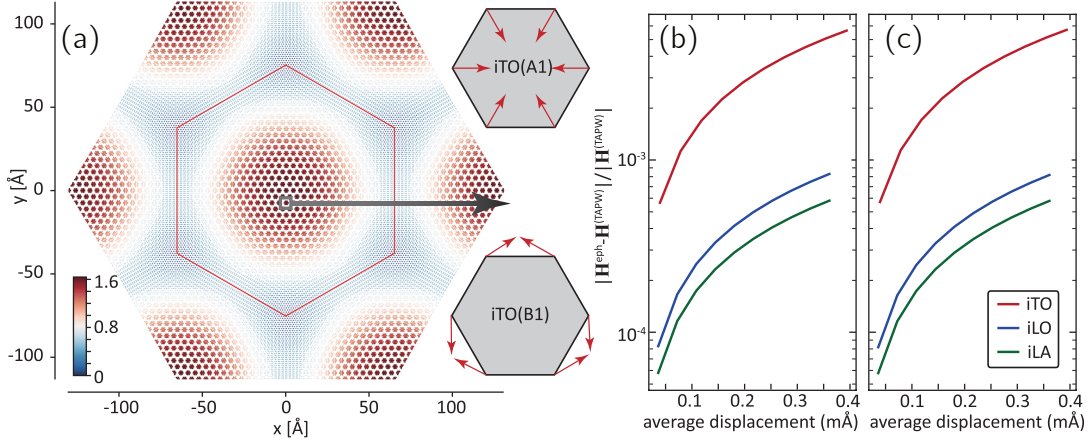


Figure 3.4: (a) Real space distortion induced by iTO moiré phonons at Γ point. The atomic vibrations are localized at the AA-stacking area and almost zero at the AB-stacking area. The red hexagon marks the Wigner-Seitz cell of MATBG. (b,c) Relative change for the Hamiltonian at K_M and Γ_M points induced by frozen phonon distortions of iTO (red), iLO (blue), and iLA (green) phonon branches.

by iTO phonons here, although the normal mode of moiré phonons may involve minimal contributions from other ingredients. As shown in panel (a) of Fig. 3.4, the real space distortion caused by Γ_M moiré phonon is localized in the AA-stacking area and obeys all D_{6h} point group symmetry, as the one found in ref. [115]. The localized vibrations behave like atomic iTO-A₁/iTO-B₁ mode which are degenerate. Moreover, the iTO moiré phonons also display strong inter-valley EPC. Within the frozen-phonon framework, the four flatbands are completely separated in energy for $|\mathbf{u}(\mathbf{r})| = 50$ mÅ, as shown in Fig. 3.5. This energy separation leads to insulating states for all integer electron fillings, which cannot be reproduced by other types of phonons.

The extended BM model considering electron-phonon coupling [116] written in the continuous limit is

$$H(\mathbf{r}) = \begin{pmatrix} H^+(\mathbf{r}) & V(\mathbf{r}) \\ V^\dagger(\mathbf{r}) & H^-(\mathbf{r}) \end{pmatrix}, \quad (3.27)$$

where $H^\pm(\mathbf{r})$ describes the original BM model ($\eta = \pm$ is the valley index) for each valley and $V(\mathbf{r})$ describes inter valley scattering. In the basis of $[A_1, B_1, A_2, B_2]$ (A/B is sublattice index and 1/2 is layer index, $V(\mathbf{r})$ can be written as

$$V(\mathbf{r}) = \begin{pmatrix} v_{11}(\mathbf{r}) & v_{12}(\mathbf{r}) \\ v_{21}(\mathbf{r}) & v_{22}(\mathbf{r}) \end{pmatrix}, \quad (3.28)$$

and

$$v_{12}(\mathbf{r}) = v_{21}(\mathbf{r}) = e^{i\mathbf{Q}\cdot\mathbf{r}} V_0 \quad (3.29)$$

$$v_{11}(\mathbf{r}) = e^{i\mathbf{Q}\cdot\mathbf{r}} (V_1 + e^{i\mathbf{G}_1\cdot\mathbf{r}} V_2 + e^{-i\mathbf{G}_2\cdot\mathbf{r}} V_3), \quad (3.30)$$

$$v_{22}(\mathbf{r}) = e^{i\mathbf{Q}\cdot\mathbf{r}} (V_1 + e^{-i\mathbf{G}_1\cdot\mathbf{r}} V_2 + e^{i\mathbf{G}_2\cdot\mathbf{r}} V_3), \quad (3.31)$$

with

$$V_0 = \begin{pmatrix} 0 & \gamma \\ \gamma & 0 \end{pmatrix}, \quad T_2 = \begin{pmatrix} \omega g' & g \\ g & \omega^{-1} g' \end{pmatrix} \quad (3.32)$$

$$V_1 = \begin{pmatrix} g' & g \\ g & g' \end{pmatrix}, \quad V_3 = \begin{pmatrix} \omega^{-1} g' & g \\ g & \omega g' \end{pmatrix} \quad (3.33)$$

In the above equation, $\mathbf{Q} = \mathbf{K}_1^+ - \mathbf{K}_2^-$, $\omega = e^{i2\pi/3}$.

Our TAPW method can help determine the effective parameters of extended BM model when performing plane wave projection, related technique details are discussed in Ref. [116] and Chapter 2. These parameters are summarized in Table 3.1: u, u' denote the intra-valley, inter-layer coupling strengths (parameters in original BM model); g, g' are inter-valley, intra-layer coupling strengths for different sublattice, respectively; γ is inter-valley, inter-layer coupling strength. In our frozen-phonon calculation, g, g' ,

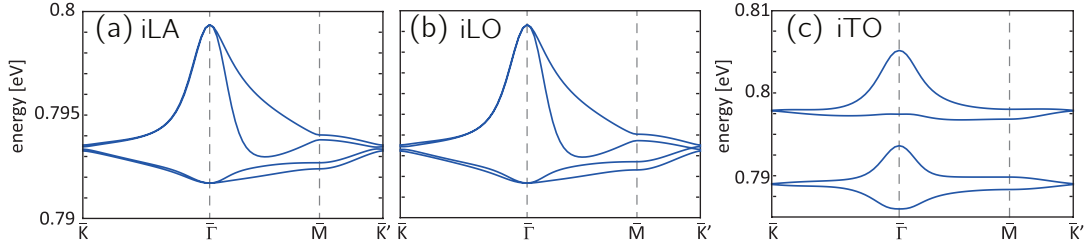


Figure 3.5: Band structure affected by frozen phonons for (a) iLO type, (b) iLA type, and (c) iTO type. The distortion field $\mathbf{u}(\mathbf{r})$ is constrained by $|\mathbf{u}(\mathbf{r})| = 50 \text{ mÅ}$. The flat bands from two atomic valleys are totally gapped out when iTO moiré phonon is frozen, which indicates strong cross valley EPC.

Table 3.1: Effective parameters extracted from TAPW method.

$ \mathbf{u} (\text{mÅ})$	$u(\text{meV})$	$u'(\text{meV})$	$g(\text{meV})$	$g'(\text{meV})$	$\gamma(\text{meV})$
5.00	85.81	103.2	0.4685	0.03758	0.01114
10.0	85.81	103.2	0.9379	0.07517	0.02228
20.0	85.81	103.2	1.873	0.1503	0.04455
30.0	85.81	103.2	2.810	0.2255	0.06681
40.0	85.81	103.2	3.746	0.3007	0.08906
50.0	85.81	103.2	4.682	0.3759	0.1111

and γ depend linearly with $|\mathbf{u}(\mathbf{r})|$, while u, u' are independent. It suggests the iTO moiré phonons only contribute inter-valley couplings, absent in the original BM model. Using these linearly dependent parameters, we can evaluate the effective EPC matrix M_0 , which is the inter-valley part of the Hamiltonian (can be built using the method described in Ref. [116] or directly read from TAPW method [53]) when $|\mathbf{u}(\mathbf{r})|$ is confined at characteristic phonon length, with second quantized phonons. We set the moiré phonon energy $\omega_0 = 150 \text{ meV}$, the mass for carbon atoms $m_c = 2.0 \times 10^{-26} \text{ kg}$, then the characteristic phonon length should be $l_p = \sqrt{\hbar/(2m_c\omega_0)} = 34.0 \text{ mÅ}$. After fixing $|\mathbf{u}(\mathbf{r})|$ to l_p , we can retrieve the normalized electron phonon coupling parameters in M_0 : $\bar{g} = 3.1882 \text{ meV}$, $\bar{g}' = 0.2569 \text{ meV}$, $\bar{\gamma} = 0.07611 \text{ meV}$.

With parameters extracted from frozen-phonon simulations, we write both the electrons and phonons in the moiré-BZ for folded TBG systems. We denote the size of moiré mini-BZ as N_m for a N -site system. The Hamiltonian of TBG bands and the linear EPC reads

$$\begin{aligned} \mathcal{H} = & \sum_{\bar{\mathbf{k}}\sigma} c_{\bar{\mathbf{k}}\sigma}^{(\text{TAPW})\dagger} h_{\bar{\mathbf{k}}} c_{\bar{\mathbf{k}}\sigma}^{(\text{TAPW})} + \sum_{\bar{\mathbf{q}}\nu} \omega_0 a_{\bar{\mathbf{q}}\nu}^\dagger a_{\bar{\mathbf{q}}\nu} \\ & - \frac{1}{\sqrt{N_m}} \sum_{\bar{\mathbf{k}}\sigma\bar{\mathbf{q}}\nu} c_{\bar{\mathbf{k}}+\bar{\mathbf{q}}\sigma}^{(\text{TAPW})\dagger} M_{\bar{\mathbf{q}}\nu} c_{\bar{\mathbf{k}}\sigma}^{(\text{TAPW})} (a_{\bar{\mathbf{q}}\nu} + a_{-\bar{\mathbf{q}}\nu}^\dagger), \end{aligned}$$

where $c_{\bar{\mathbf{k}}\sigma}^{(\text{TAPW})}$ is the column vector of electron annihilation operators with moiré momentum $\bar{\mathbf{k}}$ and spin σ in the TAPW basis (labeled by the hyper-index $(\mathbf{G}\alpha)$, where $\mathbf{G}$ is moiré reciprocal lattice vector and $\alpha \in \{A_1, B_1, A_2, B_2\}$ with A, B for atoms in the primitive cell of graphene and the subscripts for two layers) and $h_{\bar{\mathbf{k}}}$ is the electronic hopping matrix for a specific moiré momentum $\bar{\mathbf{k}}$. Together with electrons, the phonon modes are also folded into the mini-BZ, accompanied by the increase of branches. Here, $a_{\bar{\mathbf{q}}\nu}$ is the phonon annihilation operator with moiré momentum $\bar{\mathbf{q}}$ and ν labels the folded branches and ω_0 is the phonon frequency, assumed to be Einstein phonon within the moiré-BZ. The coupling matrix between electrons and phonons is denoted as $M_{\bar{\mathbf{q}}\nu}$, whose long-wavelength limit is the M_0 mentioned above.

3.4.2 Maximally coupled moiré iT0 phonons

Similar to moiré electrons in flat bands, we expect that the minimal incorporating phonons will dominate the electron-phonon coupling (EPC) effect, with decreasing contributions from other components corresponding to high-order Umklapp processes [169–172]. However, the highly localized lattice vibrations observed in Raman experiments, particularly the one at 1580 cm^{-1} , cannot be explained using this minimal in-

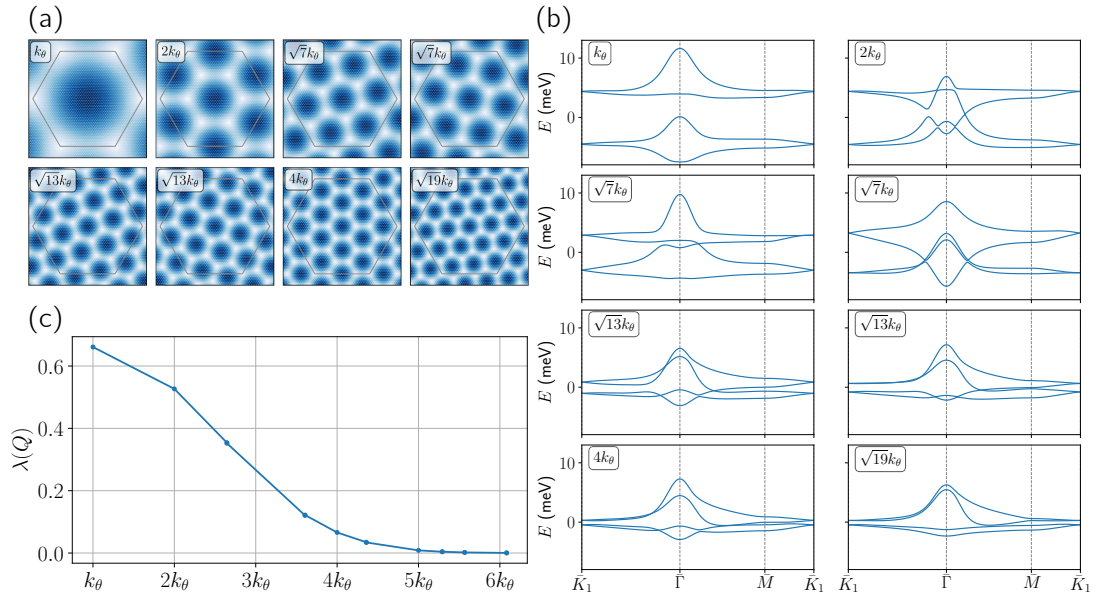


Figure 3.6: (a) Distortion strength distribution of the iTO moiré phonon modes with different orders of $\mathbf{Q}$ components on the top layer. The corresponding one on the bottom layer can be obtained through a C_{2x} operation. Only the 8 leading patterns are plotted. (b) The corresponding frozen phonon band structures calculated using TAPW method. The atomic positions are distorted by the moiré phonons field normed with the characteristic phonon length, $\sqrt{\sum_{i\alpha} \mathbf{u}^2(\mathbf{R}_{Ii\alpha})} = l_p$. (c) The (normalized) coupling strength $\lambda(|\mathbf{Q}|)$ with ZLLs shows a decaying feature when $|\mathbf{Q}|$ gets larger and become negligible when $|\mathbf{Q}| > 5k_\theta$.

corporating model. To better understand the interplay between optical moiré phonons and flat-band electrons, we need to consider higher-order components (higher-order $\mathbf{Q}$) in Eq.(3.26) for iT0 phonons.

We illustrate the real-space distortion strength induced by phonon vibrations with different orders of components in Fig. 3.6. In frozen phonon numerics, we constrain the moiré scale distortion at the characteristic phonon length l_p . Next, the effective Hamiltonian H^{TAPW} (with matrix elements denoted as $h_{\bar{\eta}\mathbf{G}'\alpha'}^{\eta\mathbf{G}\alpha}(\bar{\mathbf{k}}, \mathbf{Q})$) containing the EPC effect can be built with distorted atomic positions. Frozen phonon calculations revealed that the leading Fourier component will completely open a gap at the charge neutrality point (CNP) in the entire mBZ, while the gap near the mBZ edge will diminish as we restrict to higher-order Fourier components, supporting our previous hypothesis.

In the low-energy regime, we focus on how these phonon modes correlate with the zeroth Landau levels (ZLLs) or f orbitals in the heavy fermion model [168]. We now project the effective Hamiltonian containing EPC into the ZLL subspace,

$$h_{\eta t, \bar{\eta} t'}^{\text{zll}}(\bar{\mathbf{k}}, \mathbf{Q}) = \sum_{\mathbf{G}\alpha\mathbf{G}'\alpha'} \mathcal{L}_{\mathbf{G}\alpha}^{\eta t*}(\bar{\mathbf{k}}) h_{\bar{\eta}\mathbf{G}'\alpha'}^{\eta\mathbf{G}\alpha}(\bar{\mathbf{k}}, \mathbf{Q}) \mathcal{L}_{\mathbf{G}'\alpha'}^{\bar{\eta} t'}(\bar{\mathbf{k}}), \quad (3.34)$$

where we take advantage of the Bloch ZLL basis $f_{\bar{\mathbf{k}}, s\eta t}^\dagger = \sum_{\mathbf{G}\alpha} \mathcal{L}_{\mathbf{G}\alpha, t}^\eta(\bar{\mathbf{k}}) c_{\bar{\mathbf{k}}s\eta\mathbf{G}\alpha}^\dagger$. Then the onsite electron phonon coupling can be written down as,

$$h_{\text{on-site}}^{\text{epc}}(\mathbf{Q}) = \frac{1}{N_m} \sum_{\bar{\mathbf{k}}} h_{\eta t, \bar{\eta} t'}^{\text{zll}}(\bar{\mathbf{k}}, \mathbf{Q}). \quad (3.35)$$

We confirmed in numerics that $h_{\text{on-site}}^{\text{epc}}(\mathbf{Q})$ only contains a $\tau_x\sigma_0/\tau_y\sigma_0$ component for the A_1/B_1 mode. We denote the coefficient before the Pauli matrix as $\lambda(\mathbf{Q})$, which reflects the coupling strength between a specific Fourier component of the moiré phonon

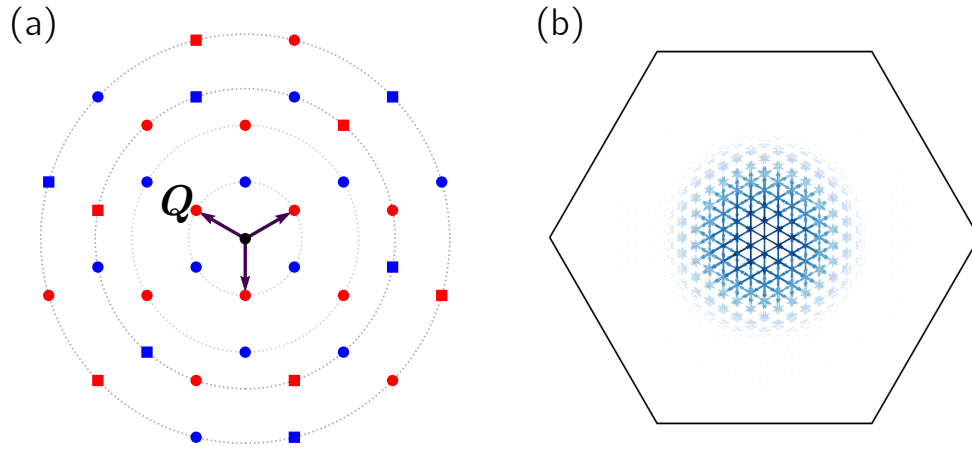


Figure 3.7: (a) The black dot in the center denotes K^+ , the black arrows connects K with three first order of Q vectors. The red dots label Q vectors in the top layer while the blue ones label the bottom layer. The C_3 connected Q vectors are labeled in the same shape. Different orders of Q vectors are grouped by grey dashed circles. (b) The real space distortion of the maximal coupled iTO moiré phonon (periodic part) with A_1 representation. The blue arrows depict the direction of atomic oscillations, and a darker colour indicates a more intense vibration. The lattice vibration has a well localized feature in the moiré scale, where the black hexagon denotes the Wigner-Seitz (WS) cell of the moiré superlattice. The center of the hexagon is the AA-stacking area and the corner of the hexagon is the AB-stacking area. The localized vibrations have atomic features.

mode and ZLLs. By linearly combining different orders of Fourier components with coefficients $\lambda(\mathbf{Q})/\lambda_t$ (where $\lambda_t = \sqrt{\sum_{\mathbf{Q}} \lambda(\mathbf{Q})^2}$ is a normalization factor), we can construct maximally coupled iTO moiré phonons. This Wannierization process allows for the selection of a particular phonon mode that contains complete correlation effects with low-energy electrons from the degenerate phonon subspace. Fig. 3.7-(b) shows that the real-space distortion induced by the Wannierized phonon modes have a well-localized feature at the AA stacking area, consistent with recent experimental Raman [127] and STM observations [154].

3.5 Conclusion

In this chapter, we first developed the truncated atomic plane wave method for low-energy moiré phonons by mapping the dynamic matrix to the tight-binding Hamiltonian. This approach is extremely efficient for solving low-frequency vibrational modes in magic-angle twisted bilayer graphene systems. For the optical phonons, we found that an in-plane transverse mode, which can lead to effective inter-valley interaction, can significantly affect the band structure if this mode is frozen. We made the Einstein phonon approximation and systematically studied the effective electron-phonon coupling of different orders. Furthermore, a most localized moiré iTO phonon can be constructed, showing a localized feature at the AA stacking area, which is consistent with recent experimental observations.

Chapter 4

Theory for magneto transport of cadmium arsenide thin films

In this chapter, we predict two topological phase transitions for cadmium arsenide (Cd_3As_2) thin films under in-plane magnetic field, taking advantage of a four-band $k \cdot p$ model and effective g factors calculated from first principles. Film thickness, growth direction and in-plane Zeeman coupling strength can all serve as control parameters to drive these phase transitions. For (001) oriented Cd_3As_2 thin films, a two dimensional Weyl semimetal phase protected by $C_{2z}\mathcal{T}$ symmetry can be realized using an in-plane magnetic field, which has recently been reported in a recent paper [184]. We then put forth two pathways to achieve in-plane anomalous Hall effects (IPAHE). By either introducing a trigonal warping term or altering the growth orientation, the emergent $C_{2z}\mathcal{T}$ symmetry can be broken. Consequently, in the clean limit and at low temperatures, quantized Hall plateaus induced by in-plane Zeeman fields become observable.

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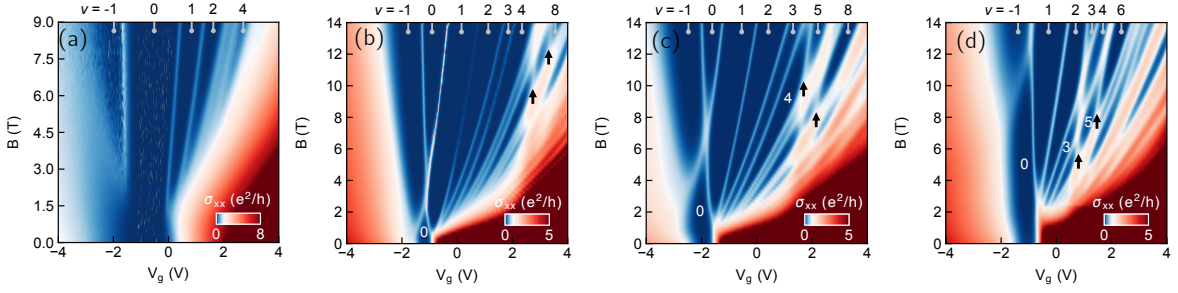


Figure 4.1: Landau fan diagram of Cd_3As_2 of different thickness (a) 14 nm (b) 18 nm (c) 19 nm (d) 22 nm. The electron band and hole band get inverted in the thicker region (18 nm - 22 nm). This figure is adapted from Ref. [4], which is copyrighted by the American Physical Society and reproduced with permission.. The Landau level diagram calculated numerically is shown in Fig. 4.4.

4.1 Introduction

Recent magnetotransport measurements [4, 184, 186] of cadmium arsenide thin films (Cd_3As_2) grown by molecular beam epitaxy (MBE) have revealed several attractive topological phases in two dimensions (2D), including a topological insulator phase and a Weyl semimetal phase. Notably, (001) grown Cd_3As_2 thin films under an in-plane Zeeman magnetic field offer a minimal model of a Weyl semimetal in 2D, where it hosts *one single* pair of Weyl nodes carrying opposite chirality, separated in the momentum space, and protected by $C_{2z}\mathcal{T}$ symmetry.

Weyl semimetals are elusive in solid-state systems [187–189]. To acquire this topological phase, either time reversal symmetry ($\mathcal{T}$) or space inversion symmetry ($\mathcal{P}$) must be broken. The first experimentally confirmed 3D Weyl semimetal [190–192], the non-centrosymmetric crystal TaAs (which breaks $\mathcal{P}$), hosts 12 pairs of Weyl nodes near the Fermi surface, and displays exotic physical phenomena including the chiral anomaly and chiral zero sounds [193, 194]. The large number of Weyl nodes near the Fermi level, however, has hindered detailed further investigations. A second route to a Weyl semimetal is to break time reversal symmetry ($\mathcal{T}$) starting with a parent

Dirac semimetal state that has large g factors. This approach has been discussed in Ref. [195, 196], and relies on an external magnetic field or magnetic dopants. The location of Weyl nodes may be controlled by the field strength, offering an experimental advantage. We point out here that the Zeeman effect and orbital effect are two major consequences of having an external magnetic field. It is not enough to only consider the Zeeman effect for *bulk* materials and ignore the orbital effect. Lastly, it was recently reported in Ref. [197] that magnetic doping remains challenging for bulk Dirac semimetals, such as Cd_3As_2 .

It is natural to ask if we can find a system hosting a minimal number of Weyl nodes, and ideally in a low-dimensional material. Several materials are predicted to host a single pair of Weyl nodes [198–204], but few have been experimentally confirmed. In 3D, while Weyl nodes are topologically stable against perturbations, external symmetry is required to protect the Weyl nodes in 2D materials. More specifically, as noted in Ref. [205, 206], a space-time inversion symmetry ($C_{2z}\mathcal{T}$) is the necessary component. The $C_{2z}\mathcal{T}$ symmetry enforces the Berry curvature to be zero everywhere in momentum space except at the band touching points that are the Weyl nodes. For example, in a Cd_3As_2 quantum well in the (001) direction, C_{2z} symmetry is inherited from the C_{4z} rotational symmetry and the product symmetry $C_{2z}\mathcal{T}$ survives as long as the external magnetic field is directed in the film plane. The 2D Weyl semimetal phase is therefore predicted to appear [207] when the quantum well subbands become inverted by the in-plane magnetic field. It is worth mentioning, that in the thin film region, the orbital effect of the magnetic field is greatly suppressed. The Zeeman effect, which is already significantly enhanced in systems with strong spin orbital coupling (SOC) [208], becomes substantial and leads to the lifting of spin degeneracy. The spin-down subbands are pushed down by the magnetic field while the spin-up subbands are lifted up. As a

result, *one* pair of unobstructed Weyl nodes is created by the Zeeman field when the in-plane magnetic field is sufficiently strong and the zero-field band gap is small. On one side, these Weyl points can be killed under a small out of plane magnetic field. On the other side, one can imagine the $C_{2z}\mathcal{T}$ symmetry can be spontaneously broken, for example, when the thin film is grown along (112) direction. As a result, the Weyl points will be gapped out and the Berry curvatures are localized near their original positions, leading the bands below the Fermi level to acquire a finite Chern number $|C| = 1$. This makes a quantized in-plane anomalous Hall effect (IPAHE) or anomalous planar Hall effect (APHE) [209–217] feasible (For the terminology used here, see ¹). These exotic quantum Hall signals emerge due to the non-trivial band topology tuned by the in-plane Zeeman field with appropriate symmetries, which is different from previous studies of so-called three dimensional quantum Hall effects [220, 221] in Cd_3As_2 thick slabs.

In this chapter, we first present the semi-classical transport theory for thin films under in-plane magnetic field and the full $k \cdot p$ Hamiltonian for Cd_3As_2 with higher order corrections in Sec. 4.2. The numerical studies of two Zeeman field induced topological phase transitions are shown in Sec. 4.3. We summarize our discovery and discuss possible experimental realization in Sec. 4.4.

¹We find that the term ‘in-plane anomalous Hall effect’ is more appropriate for the phenomena described in this chapter, as discussed in Ref. [212]. However, it’s worth noting that some papers refer to this as the ‘anomalous planar Hall effect’ [218, 219].

4.2 General theory and model Hamiltonian

4.2.1 Bulk g factor calculation

We now first review the methodology [222, 223] of the computation of effective g factors integrated in the bulk $k \cdot p$ model with strong spin orbital coupling. The full $k \cdot p$ Hamiltonian is usually expanded near a high symmetry k point,

$$H_{nn'}(\mathbf{k}) = \delta_{nn'} \left(\varepsilon_n + \frac{\hbar^2 \mathbf{k}^2}{2m} \right) + \mathbf{v}_{nn'} \cdot \mathbf{k}. \quad (4.1)$$

where n, n' is the band index and $\mathbf{v}_{nn'}$ is the velocity matrix which can be obtained through first principle calculations. This full $k \cdot p$ model should be downfolded to a much smaller Hilbert space which only contains active bands (m) near the Fermi surface, the downfolded Hamiltonian is written as:

$$H_{mm'} = \delta_{mm'} \left(\varepsilon_m + \frac{\hbar^2 \mathbf{k}^2}{2m} \right) + \mathbf{v}_{mm'} \cdot \mathbf{k} + \frac{1}{2} \sum_{\ell, ij} \left(\frac{1}{\varepsilon_m - \varepsilon_\ell} + \frac{1}{\varepsilon_{m'} - \varepsilon_\ell} \right) v_{m\ell}^i v_{\ell m'}^j k^i k^j. \quad (4.2)$$

The second term is obtained through Lowdin perturbation which describes the effective coupling between low energy subspace and high energy subspace. When the magnetic field is applied, k^i/k^j are not good quantum numbers anymore and Peierls substitution $k^i \rightarrow \tilde{k}^i = -i\partial_i + eA/\hbar$ should be applied. $\tilde{k}^i \tilde{k}^j$ do not commute but it can be separated into a symmetric part and an anti-symmetric part. The later one captures the orbital magnetic momentum of the Bloch electrons through an effective g factor tensor description,

$$g_{mm'}[\mu_B B] = -i \frac{m_e}{\hbar^2} \sum_{\ell, ijk} \left(\frac{1}{\varepsilon_m - \varepsilon_\ell} + \frac{1}{\varepsilon_{m'} - \varepsilon_\ell} \right) v_{m\ell}^i v_{\ell m'}^j \epsilon_{ijk} e_k. \quad (4.3)$$

This effective g factor tensor calculated for the bulk materials is k dependant and it integrated out the coupling between remote bands and external magnetic field. For simplicity, we made the approximation $g(\mathbf{k}) \approx g(\Gamma)$.

4.2.2 Semi-classical transport theory

We start by introducing the linear order response of the non-magnetic thin film system subjected to an in-plane magnetic field. This analysis will serve as a foundation for investigating the case of Cd_3As_2 . The semi-classical equation of motion for the Bloch electron wave packet can be formulated, taking into account the applied magnetic and electric fields [224] (we take $e = \hbar = 1$),

$$\begin{aligned}\dot{\mathbf{r}} &= \mathcal{D}^{-1}(\mathbf{v} + \mathbf{E} \times \boldsymbol{\Omega} + (\boldsymbol{\Omega} \cdot \mathbf{v})\mathbf{B}) \\ \dot{\mathbf{k}} &= \mathcal{D}^{-1}(-\mathbf{E} - \mathbf{v} \times \mathbf{B} - (\mathbf{E} \cdot \mathbf{B})\boldsymbol{\Omega}),\end{aligned}\tag{4.4}$$

where $\boldsymbol{\Omega}$ is the berry curvature, $\mathcal{D} = 1 + \mathbf{B} \cdot \boldsymbol{\Omega}$ is the volume factor, and $\mathbf{v}$ is the group velocity. The band index is neglected here. When the electrical field and magnetic field is co-planar, we assume $\mathbf{E} = (E, 0, 0)$ and $\mathbf{B} = B(\cos \theta, \sin \theta, 0)$. The linear order transverse conductivity $\sigma_{yx} = \sigma_{yx}^{(1)} + \sigma_{yx}^{(2)}$ can be solved under relaxation approximation [225–227]:

$$\begin{aligned}\sigma_{yx}^{(1)} &= \tau \int [d\mathbf{k}] \mathcal{D}^{-1} \left(-\frac{\partial f_{\text{eq}}}{\partial \varepsilon} \right) [v_x + B \cos \theta (\boldsymbol{\Omega} \cdot \mathbf{v})] \\ &\quad \times [v_y + B \sin \theta (\boldsymbol{\Omega} \cdot \mathbf{v})] \\ \sigma_{yx}^{(2)} &= \int [d\mathbf{k}] f_{\text{eq}} \Omega_z,\end{aligned}\tag{4.5}$$

where f_{eq} is the equilibrium Fermi-Dirac distribution function with the energy dispersion ε and high order contributions are ignored here. The first part $\sigma_{xy}^{(1)}$ contains traditional planar Hall response and Drude term, which scales linearly with relaxation time

τ . This contribution has been extensively studied in three dimensional topological Weyl semimetal and nodal semimetal [226, 227]. The second part $\sigma_{xy}^{(2)}$ is dissipationless and anti-symmetric which results from non-trivial Berry curvature. In experiments, these contributions to the transverse conductivity can be distinguished through taking measurements with $\mathbf{B}$ and $-\mathbf{B}$ and performing scaling analysis. In this chapter, we mainly focus on how in-plane magnetic field will contribute to anomalous Hall conductivity $\sigma_{yx}^{(2)}$. One can imagine, in the two dimensional limit, the in-plane magnetic field will not produce Lorentz force and it only interacts with Bloch electrons through effective Zeeman coupling. This phenomenon only holds in the thin film region, where the film thickness is approximately 25 nm or less, and the magnetic length is comparable to the film thickness, typically for magnetic fields below 15 T. The quantized plateaus induced by the Zeeman effect in this system differ from the previously studied 3D quantum Hall effects in Cd_3As_2 thick slabs (with dimensions on the scale of 100 nm to 200 nm, as reported in Ref. [220, 221]). In those studies, Landau levels were observed.

In this study, we present the effective g factor tensor in the bulk $k \cdot p$ model using first principle wave functions following Ref. [222, 223]. Consequently, we can simultaneously capture the in-plane magnetic field-induced corrections for band dispersion, group velocity and Berry curvature, which were previously treated as a perturbative $O(B)$ correction in Ref. [224, 228–230].

4.2.3 Low energy effective model

We first revisit the low energy effective model for bulk Cd_3As_2 , which belongs to tetragonal crystal system. The effective orbitals near the Fermi surface can be represented as $|\frac{1}{2}, \pm\frac{1}{2}\rangle$ and $|\frac{3}{2}, \pm\frac{3}{2}\rangle$ and the symmetry allowed low energy [195, 196, 231]

$k \cdot p$ Hamiltonian then reads:

$$\mathcal{H}_0(\mathbf{k}) = \epsilon(\mathbf{k}) + \begin{pmatrix} \mathcal{M}(\mathbf{k}) & Dk_- & \mathcal{A}^*(\mathbf{k}) & \mathcal{B}^*(\mathbf{k}) \\ Dk_+ & \mathcal{M}(\mathbf{k}) & \mathcal{B}(\mathbf{k}) & -\mathcal{A}(\mathbf{k}) \\ \mathcal{A}(\mathbf{k}) & \mathcal{B}^*(\mathbf{k}) & -\mathcal{M}(\mathbf{k}) & 0 \\ \mathcal{B}(\mathbf{k}) & -\mathcal{A}^*(\mathbf{k}) & 0 & -\mathcal{M}(\mathbf{k}) \end{pmatrix}, \quad (4.6)$$

where $\epsilon(\mathbf{k}) = C_0 + C_1 k_z^2 + C_2(k_x^2 + k_y^2)$, $\mathcal{M}(\mathbf{k}) = M_0 + M_1 k_z^2 + M_2(k_x^2 + k_y^2)$, $\mathcal{A}(\mathbf{k}) = Ak_- + B_1(k_x^3 - ik_y^3) + iB_2 k_+ k_x k_y - B_5 k_- k_z^2$, $\mathcal{B}(\mathbf{k}) = B_3(k_x^2 - k_y^2)k_z + iB_4 k_x k_y k_z$, $k_{\pm} = k_x \pm ik_y$. $Dk_{\pm}$ term breaks the inversion symmetry and cubic terms characterize the C_{4z} rotational symmetry. The x, y, z axis of the Hamiltonian are defined along (100), (010), (001) directions. Up to the second order, this Hamiltonian hosts two Dirac points located at $(0, 0, \pm\sqrt{-M_0/M_1})$. In bulk, single crystals of Cd_3As_2 , structures with and without an inversion center have been observed, as reported in [232–234], which demonstrate that growth and processing conditions determine the specific crystal structure. In this study, we mainly focus on the one with inversion symmetry ($Dk_{\pm} = 0$), which is based on experiments that show the preservation of inversion centers in high-quality MBE-grown Cd_3As_2 thin films on III-V substrates. This point has been carefully examined using convergent beam electron diffraction technique in Ref. [186]. Note that, the $k \cdot p$ Hamiltonian possesses a C_{∞} rotational symmetry in (001) direction without cubic terms. Furthermore, the Zeeman coupling term for the bulk $k \cdot p$ model can

be constructed [26]:

$$\mathcal{H}_{\text{Zeeman}} = \mu_B \begin{pmatrix} g_{1z}B_z & g_{1p}B_- & 0 & 0 \\ g_{1p}B_+ & -g_{1z}B_z & 0 & 0 \\ 0 & 0 & g_{2z}B_z & g_{2p}B_+ \\ 0 & 0 & g_{2p}B_- & -g_{2z}B_z \end{pmatrix} \quad (4.7)$$

where the magnetic field is chosen as $\mathbf{B} = (B_x, B_y, B_z)$ and $B_{\pm} = B_x \pm iB_y$. We adopt the following parameters for the $k \cdot p$ model, $C_0 = -0.0145$ eV, $C_1 = 10.59$ eV Å, $C_2 = 11.5$ eV Å², $M_0 = -0.0205$ eV, $M_1 = 18.77$ eV Å², $M_2 = 13.5$ eV Å², $A = 0.889$ eV Å [195]. $B_1 = 0.0153$ eV Å³, $B_2 = -0.0476$ eV Å³, $B_3 = 0.0669$ eV Å³, $B_4 = -0.0366$ eV Å³, $B_5 = 0.0668$ eV Å³, $g_{1p} = 11.6211$, $g_{2p} = 0.5876$, $g_{1z} = 10.0316$, $g_{2z} = -4.3048$ are new parameters calculated using first-principles wave-functions following the procedures described in Ref. [222, 223]. To acquire the band dispersion of Cd₃As₂ thin film grown in a general direction, denoted as z' , we rotate z axis to z' axis using the following matrix:

$$\begin{pmatrix} k'_x \\ k'_y \\ k'_z \end{pmatrix} = \begin{pmatrix} \cos \theta & -\sin \theta & 0 \\ \sin \theta \cos \phi & \cos \theta \cos \phi & -\sin \phi \\ \sin \theta \sin \phi & \cos \theta \sin \phi & \cos \phi \end{pmatrix} \begin{pmatrix} k_x \\ k_y \\ k_z \end{pmatrix}. \quad (4.8)$$

we set $(\theta, \phi) = (0, 0), (\pi/4, \arctan(\sqrt{2}/2))$ for Cd₃As₂ thin films grown along the (001) and (112) directions, respectively, which are available through MBE. The DFT calculated band structure for bulk Cd₃As₂ and the energy levels at the Γ point for 100 nm thick slabs for different orientations are shown in Fig. 4.3.

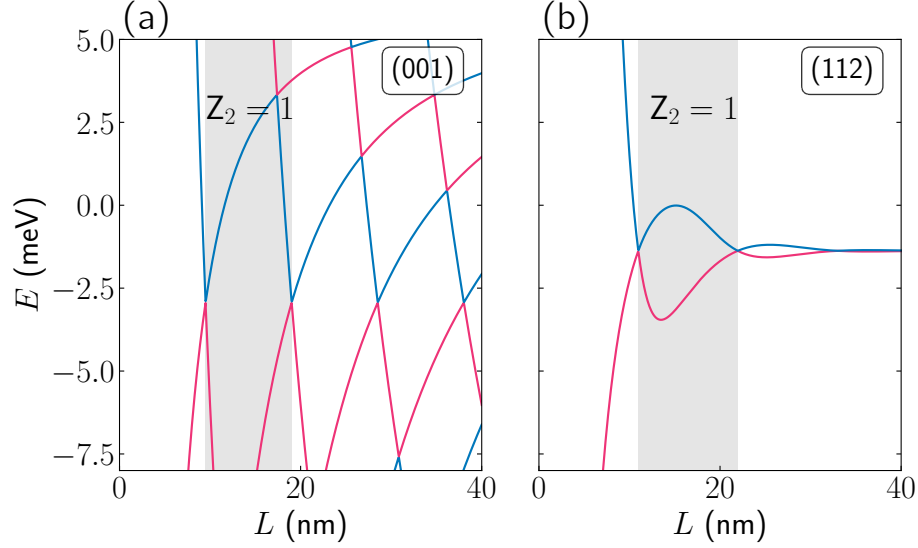


Figure 4.2: Energy levels at the Γ point near the Fermi surface versus the thin films thickness for (a) (001) thin films, (b) (112) thin films. Here, we take $D = 0$ and ignore cubic terms in Eq. (4.6). The 2D-TI phase appears in an oscillatory fashion as the function of the thickness of the thin film. We labeled the first 2D-TI ($Z_2 = 1$) region in the shadowed area, which has the best chance to be realized in experiments.

4.2.4 Critical thickness for Cd_3As_2 thin films

We emphasize that in order to achieve the in-plane Zeeman field-induced topological transition, two conditions must be satisfied: the film thickness should be sufficiently thin, and the band gap should be small. By meeting these criteria, the band inversion can occur through the application of a moderate magnetic field. Therefore, it becomes crucial to determine the critical thickness of the thin film for different growth orientations. It was first proposed in Ref. [235–237] that the electron band and hole band will oscillate with the film thickness and two dimensional topological insulator (2D-TI) phase can be obtained as a result of the quantum confinement effect. This is the case for Dirac semimetal Cd_3As_2 [231]. We start by presenting the sub-band structures of Cd_3As_2 thin films. To simulate the corresponding quantum well (QW) structure in experiments, the continuous limit that $k'_z = -i\partial_{z'}$ and open boundary condition along z'

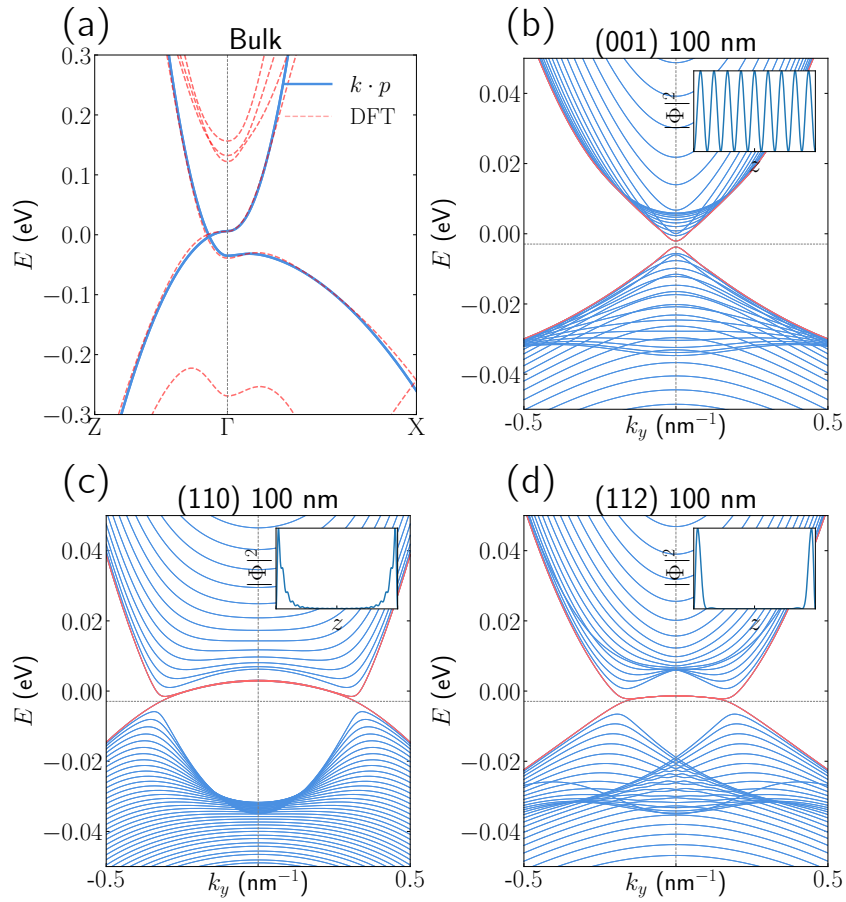


Figure 4.3: (a) DFT calculated band structure for bulk Cd_3As_2 and the corresponding one using $k \cdot p$ model. (b)-(c) Calculated energy bands of 100 nm slabs grown along different orientations. (b)(001) direction (c) (110) direction and (d) (112) direction. The insets plot the wavefunction distribution along z direction.

directions are applied. The Hamiltonian $\mathcal{H}(\mathbf{k}') = \mathcal{H}(k'_x, k'_y, -i\partial_{z'})$ can then be expanded using a plane-wave basis

$$\psi_n(z') = \sqrt{\frac{2}{L}} \sin \left[\frac{n\pi}{L} \left(z' + \frac{L}{2} \right) \right], \quad -L/2 < z' < L/2, \quad (4.9)$$

where L is the thickness of the thin film and n is taken up to 50. As shown in Fig. 4.2, the electron band and hole band oscillate, which allows us to obtain the 2D TI phase by tuning the thin film thickness. We first focus on the thin films grown along (001) direction. By direct inspection of Eq. (4.6) (set $D = 0$ and ignore cubic terms), we can easily solve for the energy levels at Γ point with $E_n = C_0 + M_0 + (C_1 + M_1)(n\pi/L)^2$, $H_n = C_0 - M_0 + (C_1 - M_1)(n\pi/L)^2$. The critical thickness L_{cn} should satisfy $E_n = H_n$. Then we get $L_{cn} = n\pi/\sqrt{-M_0/M_1}$ and the same analysis can be performed for (112) grown thin films. For (001) thin films, $L_{c1} = 10$ nm, $L_{c2} = 19$ nm, for (112) thin films, $L_{c1} = 11$ nm, $L_{c2} = 21$ nm. We labeled the first 2D-TI region in Fig. 4.2 and the numerical result is consistent with recent transport experiment [4]. Among different thicknesses, a thin film grown around 20 nm has the highest probability of realizing Zeeman field induced effect. In the upcoming discussion, we will delve into the details of this phenomenon.

4.3 Zeeman-field induced physical effects

In this section, we demonstrate that Cd_3As_2 thin films can manifest rich topological phase diagrams under the influence of an in-plane magnetic field. The Zeeman effect serves to lift the spin degeneracy of the sub-bands, leading to their inversion when the in-plane magnetic field is sufficiently strong. For thin films grown along the (001) direction, a single pair of Weyl points can be stabilized by $C_{2z}\mathcal{T}$ symmetry. Once this

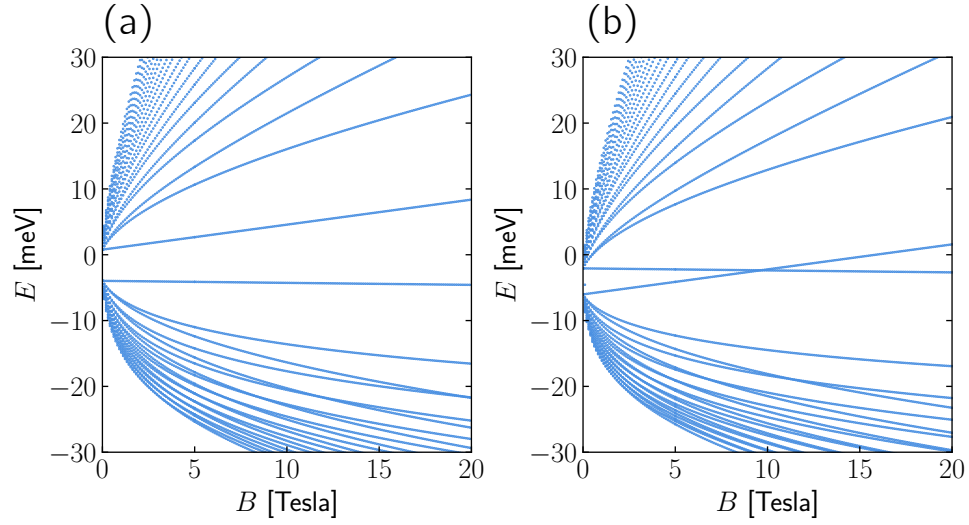


Figure 4.4: Calculated Landau levels for (001) grown thin films when an out of plane B field is applied. (a) The uninverted region. (b) The inverted region. The corresponding parameter settings will be discussed in Chapter 5.

symmetry is broken, the emergence of the in-plane quantum anomalous hall effect can be anticipated. Conversely, for thin films grown along the (112) direction and near a critical thickness, a topological gap remains robust when an in-plane Zeeman field is introduced.

4.3.1 Two dimensional Weyl semimetal phase

We start with a general analysis for (001) grown Cd_3As_2 thin films under in-plane magnetic field. We can expect the orbital effect (which results in Landau Levels) and Zeeman effect to co-exist when the field is switched on. We first estimate the magnetic length $\ell_B = \sqrt{\hbar/eB} = 25.6/\sqrt{B[\text{T}]}[\text{nm}]$ is around 10 nm for 10 T which is comparable with the thin film thickness. The cyclotron motion of the electrons along (001) direction is greatly suppressed by the quantum confinement effect. On the other side, the Zeeman effect leads to the spin splitting and the energy levels at Γ point can be mod-

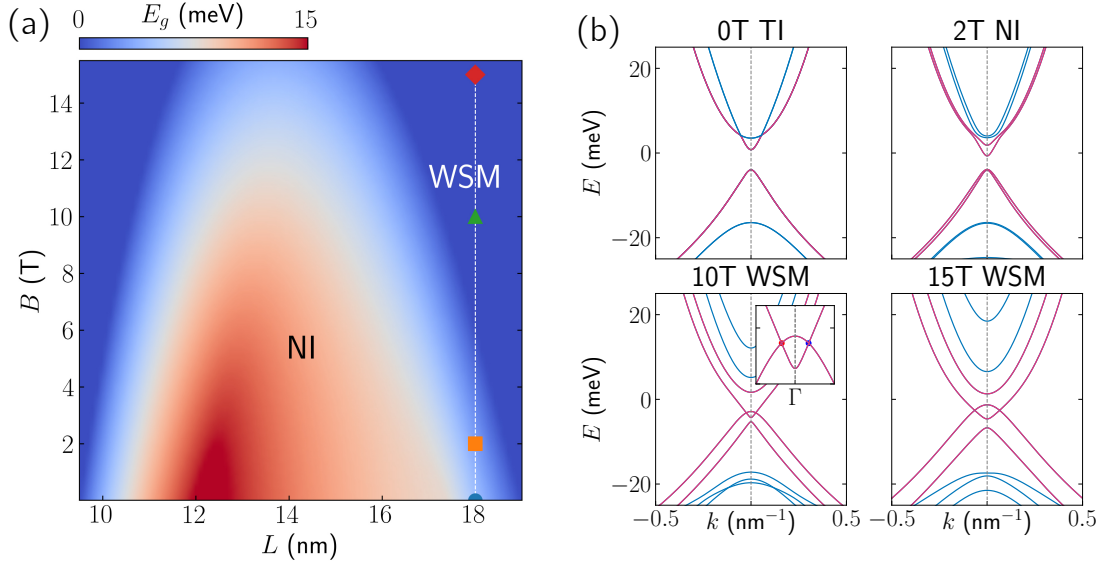


Figure 4.5: (a) Topological phase diagram for (001) grown Cd_3As_2 thin films under in-plane magnetic field B . The colorbar labels the calculated band gap of the thin films at the Γ point. We focus on the 9 nm to 19 nm-thick thin films which are in the 2D-TI phase when there is no magnetic field. The normal insulator (NI) phase and Weyl semimetal (WSM) phase are labeled in different colored regions. We show the most possible thickness to realize WSM phase is close to the critical point (19 nm). For an 18 nm thin film, we take a line cut on the phase diagram to illustrate different phases. The half-circle denotes the 2D-TI phase (0 T), the square denotes the normal insulator phase (2 T), the triangular is the WSM phase (10 T) and the diamond is the WSM phase (15 T). (b) Band structures calculated along the direction perpendicular to the magnetic field for 18 nm (001) grown thin films under in-plane magnetic field. Zeeman effect and orbital effect are all considered. Four central bands close to the Fermi level are depicted in red while other bands are in blue. When $B = 0$ T, the system is in a 2D-TI phase, all bands are doubly degenerate. When $B = 2$ T, the system is a normal insulator. When $B = 10, 15$ T, the system is in a WSM phase. The inset shows the band dispersion of the 2D WSM, but within a lower energy range. Two Weyl nodes are separated in the momentum space and a stronger in-plane magnetic field can push them away from each other.

ified by a magnitude of $g\mu_B B$. It is noteworthy that the energy scale of the Zeeman splitting is typically small in comparison to the large band gap found in conventional semiconductors. However, the band gap in these topological thin films is readily tunable, rendering the Zeeman effect particularly significant. As a result, the topological gap of the 2D-TI will be inverted. One pair of Weyl nodes appear at the band crossing point and they are protected by the new emergent $C_{2z}\mathcal{T}$ symmetry. This new symmetry guarantees the Berry curvature to be zero everywhere in the momentum space except the Weyl nodes and the Berry phase integrated along the path enclosing the Weyl nodes must be $\pm\pi$.

We confirmed above analysis using numerical calculations. We first assume $\mathbf{B} = (B_x, B_y, 0)$, $B_x = B \cos \varphi$, $B_y = B \sin \varphi$ and set $D = 0$, $B_{1-5} = 0$ for simplicity, the full Hamiltonian incorporating in-plane magnetic field should be:

$$\mathcal{H}_B = \mathcal{H}_0 \left(k'_x + \frac{z'}{\ell_{B_y}^2}, k'_y - \frac{z'}{\ell_{B_x}^2}, -i\partial_{z'} \right) + \mathcal{H}_{\text{Zeeman}}, \quad (4.10)$$

where Landau gauge and Peierls substitution are applied to fully consider orbital effect. Then the magnetic Hamiltonian $\mathcal{H}_B$ can be expanded under the plane-wave basis $\psi_n(z')$ defined in Eq. (4.9). The computed phase diagram illustrating the band gap as a function of the in-plane magnetic field B and the film thickness L is presented in Fig.4.5-(a). Notably, when the film thickness approaches the critical point L_c , even a moderate magnetic field can induce a phase transition from a normal insulator (NI) to a Weyl semimetal (WSM). For instance, consider an 18 nm thin film, as shown by the line cut in Fig.4.5-(a) and the corresponding band structures in Fig.4.5-(b). In the 2D-TI phase without an applied magnetic field, all sub-bands are doubly degenerate. With a sufficiently strong in-plane magnetic field (around 10 T), a pair of Weyl nodes emerges in the direction perpendicular to the magnetic field. We also explore the role

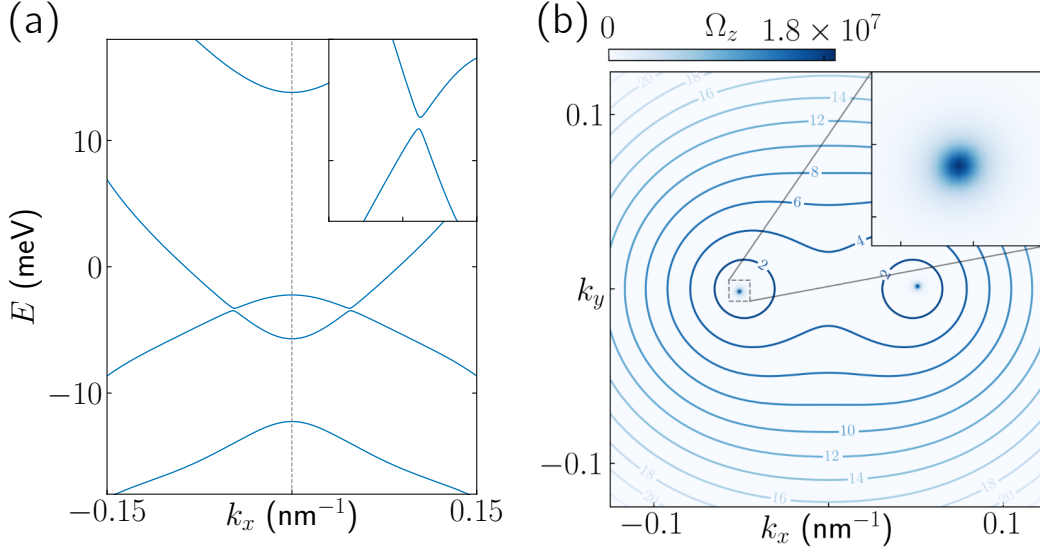


Figure 4.6: (a) The band structure for the 2D Weyl semi-metal phase ($B_y = 15$ T) after integrating the trigonal warping term. ($R_1, R_2, R_3, R_4 = 100$ eV $\text{\AA}^3$) A small non-trivial gap is opened, as shown in the inset. (b) Calculated Berry curvature $\Omega_z(\mathbf{k})$ for all bands below Fermi level in the momentum space. The trigonal warping results in the singular behaviour of the Berry curvature. As depicted in the inset, the berry curvature concentrates near the original Weyl points. The contours for the band gap is also depicted in units of meV.

of orbital effects in our calculations. Both Zeeman and orbital effects are accounted for in the sub-band structures depicted in Fig.4.5-(b). Our findings indicate that the orbital effect is suppressed in this thin film due to quantum confinement, meaning no emergence of Landau levels. Nonetheless, the orbital effect acts to spatially separate the Weyl nodes in momentum space. This 2D Weyl semi-metal phase is stable against inversion symmetry broken term $Dk_{\pm}$ and cubic terms obeying C_{4z} rotational symmetry in Eq.(4.6), because $C_2\mathcal{T}$ symmetry survives after considering these perturbation [205].

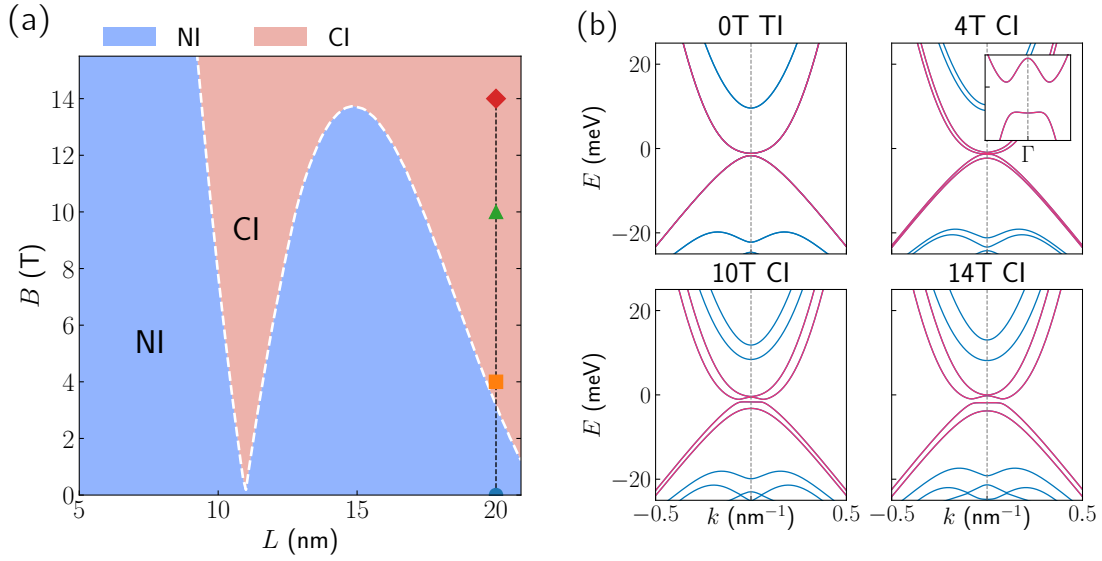


Figure 4.7: (a) Topological phase diagram for (112) Cd_3As_2 thin films under in-plane magnetic field. The normal insulator (NI) phase and Chern insulator (CI) phase are labeled in different colored regions. The white dashed curve represents the minimal in-plane magnetic field to realize CI phase at each thickness. We show the most possible thickness to realize CI phase is close to the critical point (21 nm). For a 20 nm thin film, we take a line cut on the phase diagram to illustrate the different phases. The circle denotes the 2D-TI phase (0 T), the square (4 T), the triangular (10 T) and the diamond (14 T) are all in the CI phase but with increasing topological gaps. (b) Calculated band structures along k'_y for a 20 nm (112) grown Cd_3As_2 thin film under different in-plane magnetic fields. Four bands close to the Fermi surface are labeled in red while other remote bands are in blue. When $B = 0$ T, it is in a 2D-TI phase, all bands are doubly degenerate. When $B \geq 14$ T, the system is in a CI phase. The topological gap persists and is enhanced by the in-plane magnetic field.

4.3.2 In-plane anomalous Hall effect

The conventional quantum Hall effect arises due to the formation of Landau levels and the breaking of time-reversal symmetry in the presence of an external magnetic field. A quantized Hall conductance can also manifest in a specific class of materials, even in the absence of an applied magnetic field. This occurs when time-reversal symmetry is spontaneously broken, either by magnetic dopants [32], intrinsic magnetic order [34], or moiré potentials [14, 35, 238, 239]. This phenomenon is termed the quantum anomalous Hall (QAH) effect [7]. As discussed in the previous section, the orbital effects of an in-plane magnetic field are suppressed in topological thin films with strong spin-orbit coupling (SOC). Concurrently, an enhanced Zeeman effect can induce the inversion of a pair of electron and hole band when the field is sufficiently strong. As a result, one could anticipate the emergence of a Chern insulator phase originating from the Zeeman effect upon the breaking of certain symmetries [209–212], an effect which is likewise *anomalous*.

The key principle for achieving a CI with an in-plane magnetic field in a 2D WSM we elaborate is to break the $C_{2z}\mathcal{T}$ symmetry. Upon the inclusion of symmetry-breaking terms, specifically trigonal warping [240]. For the case of (001) grown Cd_3As_2 , it can be written as:

$$\mathcal{H}_{\text{tri}}(\mathbf{k}) = \begin{pmatrix} 0 & 0 & (R_3 + iR_4)k_+k_{\parallel} & (R_1 + iR_2)k_+k_{\parallel} \\ 0 & 0 & (R_1 - iR_2)k_-k_{\parallel} & (-R_3 + iR_4)k_-k_{\parallel} \\ (R_3 - iR_4)k_-k_{\parallel} & (R_1 + iR_2)k_+k_{\parallel} & 0 & 0 \\ (R_1 - iR_2)k_-k_{\parallel} & (-R_3 - iR_4)k_+k_{\parallel} & 0 & 0 \end{pmatrix}, \quad (4.11)$$

where $k_{\parallel} = k_x^2 + k_y^2$ and R_1, R_2, R_3, R_4 are real numbers. After integrating the trigo-

nal warping in the minimal 2D WSM model, a non-trivial topological gap appears, as shown in Fig. 4.6-(a). The Berry curvature $\Omega_z(\mathbf{k})$ shows a localized distribution near the initial positions of the Weyl nodes, as illustrated in Fig. 4.6-(b). The corresponding integral of $\Omega_z(\mathbf{k})$ becomes quantized to 2π . From an experimental perspective, this kind of warping effect can be introduced in (001) grown thin films through introducing C_3 symmetric moiré potential [53, 241–246] through gate engineering [247] or proximity effect [248]. This will be discussed further in Chapter 5.

Then we focus on a realistic scenario to realize Zeeman field induced quantized IPAHE in Cd_3As_2 . We further observe that thin films of Cd_3As_2 grown along the (112) direction, which corresponds to the (111)-axis in an approximate cubic unit cell, offer an exceptional platform. It is noteworthy that unlike the (001)-grown films, those grown along the (112) direction possess only an approximate C_3 rotational symmetry. This naturally breaks the $C_{2z}\mathcal{T}$ symmetry when an in-plane magnetic field is applied. To account for this type of symmetry breaking, we take the cubic terms in Eq. (4.6) into consideration. The topological phase diagram for these (112)-grown Cd_3As_2 thin films under an in-plane magnetic field is presented in Fig. 4.7-(a). Note that, in the unrotated coordinate, $B_x + B_y + 2B_z = 0$ should be satisfied. If φ captures the angle between the magnetic field and k'_x axis, we get $B_x = (\sqrt{2}/6 + \sqrt{2}/3)B \cos \varphi + 1/\sqrt{3}B \sin \varphi$, $B_y = -(\sqrt{2}/6 + \sqrt{2}/3)B \cos \varphi + 1/\sqrt{3}B \sin \varphi$, $B_z = -1/\sqrt{3}B \sin \varphi$. The white dashed line in the diagram indicates the minimum magnetic field magnitude required to drive the system into the CI phase. The most favorable region for this transition is close to the critical thickness where the band gap is nearly zero. To illustrate the Zeeman effect-induced phase transition, we take a 20 nm thin film and set $\varphi = \pi/15$ as an example. In the absence of a magnetic field, the thin film behaves as a 2D TI, as shown in Fig. 4.7-(b). Application of a small in-plane magnetic field leads to band inversion. Unlike

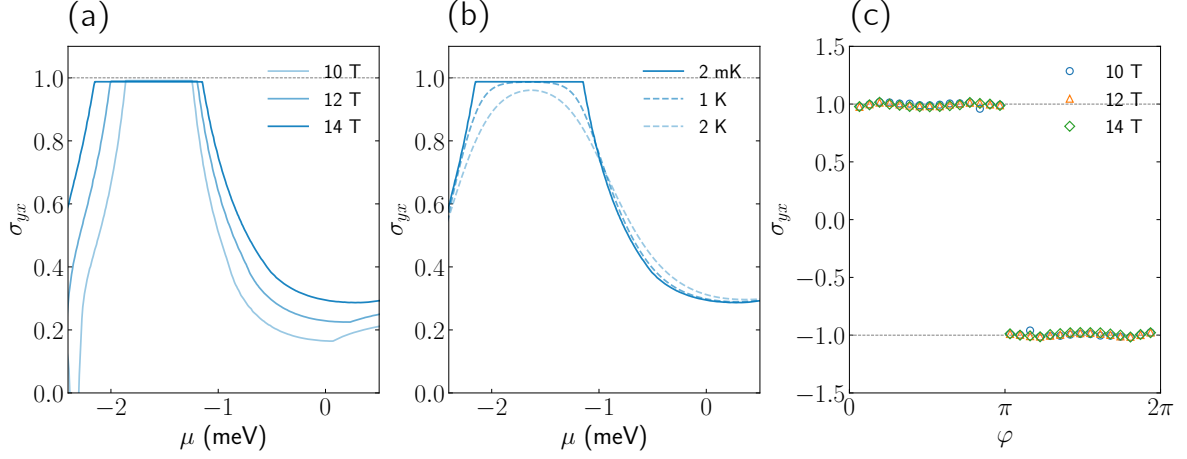


Figure 4.8: (a) Calculated Hall conductance $|\sigma_{yx}|$ versus the chemical potential at 2 mK for a 20 nm (112) grown thin film, with different magnitudes of applied in-plane magnetic fields ($\varphi = \pi/15$). (b) Calculated Hall conductance at different temperatures versus the chemical potential for a 20 nm (112) grown thin film applied with a 14 T in-plane magnetic field. The quantized plateaus are stable at low temperatures. (c) Quantized anomalous Hall conductance (chemical potential is set in the gap) versus the angle between the in-plane magnetic field B and k'_x . B is set at 10/12/14 T. It is clear that $\sigma_{yx}(B) = -\sigma_{yx}(-B)$.

the 2D WSM phase observed in (001)-grown thin films, the avoided band crossing is highlighted in Fig. 4.7-(b). Increasing the magnitude of the in-plane magnetic field enlarges the topological gap. When $B \geq 14$ T, a sizable gap opens, facilitating the quantized IPAHE.

We now investigate the impact of in-plane magnetic field strength and temperature on the quantized Hall conductance plateau in a 20 nm thin film using Kubo formula. In numerics, we preserve 32 subbands, use a 1500×1500 k -mesh and truncate the momentum space in a square region ($-0.8 \text{ nm}^{-1} \leq k'_x, k'_y \leq 0.8 \text{ nm}^{-1}$). The calculated plateau for Hall conductance is close to quantized. As depicted in Fig. 4.8(a)-(b), the width of the plateau increases with the application of a stronger magnetic field $B \geq 10$ T and is stabilized at an ultra-low temperature $T \leq 1$ K. We here emphasize that the IPAHE is distinct from the conventional planar Hall effect. Specifically, the Hall

conductance for IPAHE is an odd function of the in-plane magnetic field, as opposed to an even function in the conventional case [212, 218, 219]. This is also confirmed in our numerics that the sign of the Hall conductance will be changed by varying the direction (varying φ) of the in-plane magnetic field, as shown in Fig. 4.8. We further notice that a small deviation from inversion symmetry may cause noteworthy Berry curvature dipole distribution which hints non-linear planar Hall effect [229, 249] can co-exist with the quantized IPAHE. The breaking of inversion symmetry can be achieved through using specific growing methods or applying a vertical electrical field [208]. This intriguing phenomenon will be the subject of future studies.

4.4 Conclusion

In this chapter, we conduct a comprehensive investigation of Zeeman-field induced topological phase transitions in Cd_3As_2 thin films. Unlike previous studies [195, 196] that mainly focus on the Zeeman effect in *bulk* materials, we highlight the critical role of Zeeman splitting in thin films, especially when coupled with quantum confinement effects. Our analysis reveals that in thin films grown along the (001) direction, the orbital effects of an in-plane magnetic field $B \leq 15$ T are negligible, and the Zeeman effect becomes the dominant factor driving topological phase transitions. For (001)-oriented films, we predict the emergence of a 2D Weyl semimetal (WSM) phase, which is stabilized by $C_{2z}\mathcal{T}$ symmetry. Furthermore, the positions of the Weyl nodes can be manipulated through the direction and magnitude of the applied in-plane magnetic field. Once the $C_{2z}\mathcal{T}$ symmetry is broken due to trigonal warping, it will give rise to Chern bands. On a parallel note, for thin films grown along the (112) direction where $C_{2z}\mathcal{T}$ is obstructed from geometry, we foresee the possibility of observing an IPAHE at

low temperatures and in the clean limit, under a strong magnetic field. Our approach for achieving these phase transitions is generally applicable to MBE-grown topological thin films characterized by strong spin-orbit coupling, large effective g -factors, and appropriate symmetries [211, 212].

Chapter 5

Emergent band topology in artificial moiré lattices

We have discussed moiré effects in twisted bilayer graphene in previous chapters of this dissertation. It is natural to ask, can we create a moiré pattern without twist? The answer is yes. The most important aspect of moiré materials is the creation of a superlattice structure, which causes the bands to fold and flatten. Furthermore, band inversion can also occur if the material properties are controlled through specific engineering, such as an applied displacement field. In this chapter, we explore the emergent band topology of two models under an external moiré potential: Bernal bilayer graphene and the BHZ model. These models both have strong connections with our previous discussions.

The contents of this chapter are based on an unpublished manuscript “Artificial moiré engineering for an ideal BHZ model, W.Miao, A. Rashidi, X. Dai.”.

5.1 Introduction

Moiré patterns with large periods emerge when layers of van der Waals materials are stacked with a twist or lattice mismatch. These patterns modify energy bands, leading to phenomena such as novel superconductivity, correlated insulators, and the (fractional) quantum anomalous Hall effect. However, inhomogeneities from lattice relaxation and strain are common in these twisted nanodevices, posing significant challenges.

The moiré potential from interlayer coupling is crucial for obtaining flat bands, rather than the twist itself [244, 245]. A large-period potential that downfolds the original bands and opens gaps at the zone boundary is what matters. These potentials create flat mini-bands, enhancing electron-electron interactions and allowing for engineered band topology.

This introduces the concept of artificial moiré engineering. By utilizing contemporary technologies, we can create designed periodic potentials [247, 250] to modulate the energy bands of two-dimensional electronic systems. Unlike twisted systems, artificial lattices provide flexible control over lattice patterns, symmetry, and potential strength, offering new degrees of freedom for designing band structures. This approach applies not only to van der Waals multilayers but also to semiconductor thin films grown by molecular beam epitaxy [250]. One method of creating such an artificial potential is using a twisted hexagonal boron nitride (h-BN) substrate [251], where intrinsic charge redistribution generates an external superlattice potential. Additionally, techniques such as dielectric engineering, which involves creating specific patterns [247, 250] during the nanofabrication process, can also be employed for this purpose.

Previous theoretical studies have shown the superlattice potentials can engineer

the topology of a single massive Dirac fermion while in the case of massless Dirac fermion, a series of satellite Dirac fermions appear [241, 242]. More recently, people found that the quantum anomalous Hall (QAH) effect is available in superlattice gated bernal bilayer graphene [243] when the spin-valley polarized states are favored by the enhanced electron electron interaction. Beyond these setups based on van der Waals materials, the superlattice effects on the surface states of the topological insulator (TI) thin films have also been studied in Ref. [252].

In this chapter, we will primarily discuss the moiré superlattice effect on two models, one is the Bernal Bilayer Graphene (BLG) and one is Bernevig-Hughes-Zhang (BHZ) model. These two models both obey $\mathcal{PT}$ symmetry, and the later one was first proposed to describe the quantum spin Hall effects in HgTe/CdTe hetero-structures. The BHZ model is also applicable to Cd_3As_2 under appropriate thickness. One can envision that a spin-valley-polarized state or a spin-polarized state, which spontaneously breaks time-reversal symmetry, could become the ground state when the mini-bands become flat. This would enable the quantum anomalous Hall effect in such a setup, as shown in Fig. 5.1. To investigate this, we adopt a two step strategy. We first examine the single particle topological phase diagram to diagnose the mini-band topology and find the optimal parameter regions where the band width is small, to assess whether the quantum anomalous Hall effect can be stabilized. We then confirm the latter argument through unconstrained Hartree-Fock (UHF) calculations to account for the electron-electron interaction effects. Our numerics show that in the moiré BHZ model the quantum anomalous Hall insulator phase and anomalous Hall metal phase can both appear in the experimental parameters region.

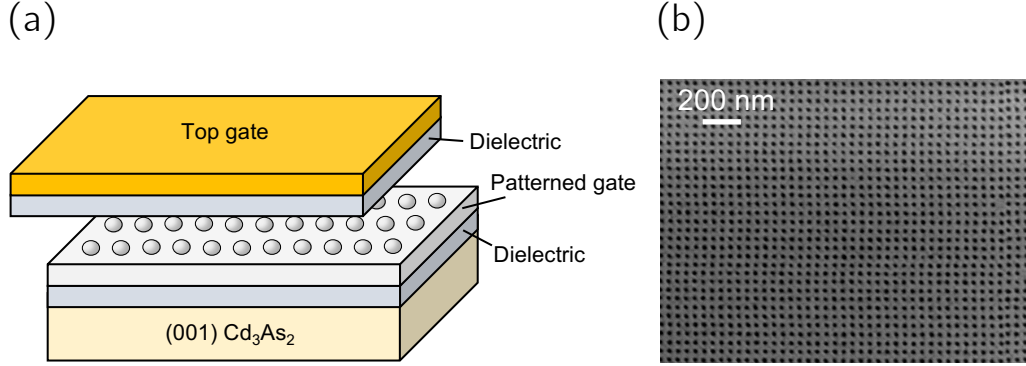


Figure 5.1: A proposed experimental device architecture in which a patterned gate metal creates a moiré potential in the Cd_3As_2 thin film. (a) illustration of the double gated device. The patterned gate sets the Fermi level in the film while the top gate controls the potential under the holes, generating an effective moiré superlattice. (b) A scanning electron microscope image of a patterned Ru gate metal realized by electron beam lithography and dry etching. The holes are around 20 nm in diameter while the pitch size is 45 nm.

5.2 Model Hamiltonian

5.2.1 Bernal bilayer graphene

The case for twisted bilayer graphene is thoroughly examined in the second chapter of this thesis. We first present the low-energy effective Hamiltonian of Bernal bilayer graphene (BLG) at the $\mathbf{K}/\mathbf{K}'$ valley [243, 253],

$$H_0 = \hbar v_F \left(\eta k_x \rho_0 \sigma_x + k_y \rho_0 \sigma_y + \frac{t}{2} (\rho_x \sigma_x - \rho_y \sigma_y) + V_0 \rho_z \sigma_0 \right), \quad (5.1)$$

where ρ and σ are Pauli matrices representing the layer and A/B sublattice degrees of freedom, respectively. The parameter $\eta = \pm 1$ captures the $\mathbf{K}/\mathbf{K}'$ valley degree of freedom. The system also exhibits $\text{SU}(2)$ spin symmetry because the spin-orbit coupling is negligible and can be ignored. Here, v_F denotes the Fermi velocity, t describes the

effective interlayer hopping, and V_0 represents an effective potential generated by an external vertical electric field.

Unlike monolayer graphene, where the low-energy Hamiltonian describes massless Dirac fermions, Bernal bilayer graphene exhibits two quadratically touching bands at the $\mathbf{K}$ valley. Moreover, applying a vertical electric field can tune the gap in this system. Note that time-reversal symmetry is broken if only one valley is considered, allowing the definition of a valley Chern number.

5.2.2 BHZ hamiltonian

The BHZ Hamiltonian serves as the minimal model to capture the transition from a normal insulator to a two-dimensional topological insulator [30]. This Hamiltonian can be decomposed into two parts: a 2×2 block describing the orbital degree of freedom and its time-reversal conjugate,

$$\mathcal{H}_{\text{BHZ}} = H_0 \oplus \mathcal{T}^{-1} H_0 \mathcal{T} = H_0(\mathbf{k}) \oplus H_0^*(-\mathbf{k}) \quad (5.2)$$

$$H_0 = C_1 \mathbf{k}^2 \sigma_0 + (\delta/2 + M_1 \mathbf{k}^2) \sigma_z + A(k_x \sigma_x - k_y \sigma_y). \quad (5.3)$$

In the above equations, δ, C_1, M_1 and A are specific material parameters at different thicknesses, and σ represents the three Pauli matrices describing the orbital degree of freedom. δ characterizes the corresponding band gap and its sign represents whether the system is in a quantum spin Hall region or a normal region. This 2×2 block can also be considered as the continuous version of famous Qi-Wu-Zhang model. In the case of Cd_3As_2 , this degree of freedom represents s or p orbitals and we take the following parameters downfolded from the original model $A = 0.889 \text{ eV } \text{\AA}$, $C_1 = 11.5 \text{ eV } \text{\AA}^2$,

$M_1 = -13.5 \text{ eV } \text{\AA}^2$. The corresponding Landau Level (LL) diagrams are depicted in Fig. 4.4. When δ and M_1 takes different sign, the zeroth LL will get inverted which hints it is in the two dimensional TI region in experiments.

The full BHZ model has inversion symmetry ($\mathcal{P}$) and time-reversal symmetry ($\mathcal{T}$) and the horizontal mirror plane ($\mathcal{M}_z$) which lead to band degeneracy everywhere. However, the time-reversal symmetry can be manually broken by considering only H_0 , and the Chern number can be defined for each spin sector. This can also be understood as the mirror Chern number defined for different mirror eigenvalue. In this chapter, we will use spin Chern number to describe the corresponding topological nature. H_0 can also be understood as a massive Dirac fermion when taking long wavelength limit $k \rightarrow 0$. Upon adding a superlattice potential, the bands may become flattened, and the electron-electron interaction can directly result in the breaking of time-reversal symmetry. The moiré potential strength and the band gap of the material can also be well designed through controlling gate voltage and the thickness of the thin film, which makes band topology engineering feasible.

5.2.3 Real space moiré potential

Generating an effective and controllable moiré superlattice potential is still a challenging task. However, several carefully designed experimental setups have demonstrated the existence of this potential. One such setup involves dielectric engineering through the creation of a patterned gate, as illustrated in Fig. 5.1. Motivated by these developments, we now present the general form of a superlattice moiré potential in real space by examining the original BM model proposed for twisted bilayer graphene:

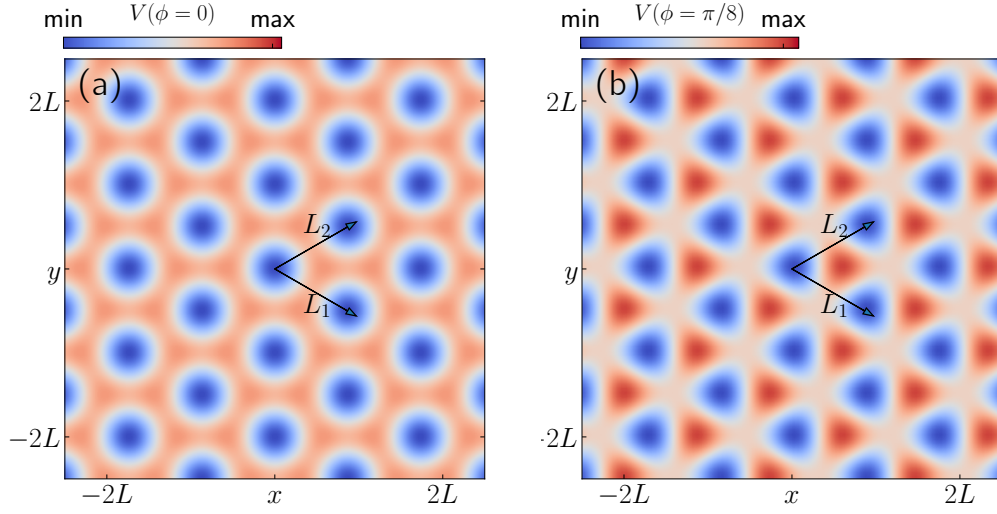


Figure 5.2: Real space distribution of the moiré potential strength $V(\mathbf{r})$ with different parameters. (a) The potential has an additional C_{6z} rotational symmetry when $\phi = 0$ (b) The potential has a C_{3z} rotational symmetry when $\phi = \pi/8$. The maximum and minimum of the strength of the potential forms two sets of triangular lattice.

$$V(\mathbf{r}) = \sum_{\mathbf{G}} V(\mathbf{G}) e^{i\mathbf{G} \cdot \mathbf{r}}, \quad (5.4)$$

where $\mathbf{G}$ is the moiré reciprocal lattice and obviously this potential obeys the translational symmetry of the moiré period. This potential is a scalar potential that does not couple with the spin and orbital degrees of freedom. This is reasonable in the case of (001) grown high-quality Cd_3As_2 thin films where surface states do not exist. However, in the case of BLG and the Bi_2Se_3 thin film family, the moiré potential exhibits layer dependence, making this scalar potential an oversimplification of the real scenario. For example, the two layers of graphene or the two surfaces of Bi_2Se_3 experience different strengths of potential. Hence, the potential should include a term that couples with ρ_z (the layer degree of freedom). We first examine the scalar case that the system obeys a

three fold rotational symmetry (C_{3z}) which has a form,

$$V(\mathbf{r}) = 2V_0 \sum_{i=1,2,3} \cos(\mathbf{G}_i \cdot \mathbf{r} + \phi), \quad (5.5)$$

where $\mathbf{G}_1 = \frac{4\pi}{\sqrt{3}L} \left(\frac{1}{2}, -\frac{\sqrt{3}}{2} \right)$, $\mathbf{G}_2 = \frac{4\pi}{\sqrt{3}L} \left(\frac{1}{2}, \frac{\sqrt{3}}{2} \right)$, $\mathbf{G}_3 = -\mathbf{G}_1 - \mathbf{G}_2$, and L is the length of the real-space moiré lattice vector. Typically in experiments, L is on the scale of 10-100 nm, and V_0 is on the scale of 10 meV. This simple moiré potential preserves the first shell of the Fourier components and has C_{3z} and time-reversal symmetry (with additional C_{6z} symmetry if $\phi = 0$). V_0 describes the strength of the potential, while ϕ corresponds to its shape, as depicted in Fig. 5.2. For the case of BLG or TI thin film, the moiré potential shall has a form of [253],

$$V(\mathbf{r}) = 2D_0 \left(\frac{1+\alpha}{2} \rho_0 + \frac{1-\alpha}{2} \rho_z \right) \sigma_0 \sum_{i=1,2,3} \cos(\mathbf{G}_i \cdot \mathbf{r} + \phi). \quad (5.6)$$

and the parameter α is designed to capture the difference in moiré potential experienced by different layers or surfaces and D_0 represents the amplitude of the displacement field.

The full Hamiltonian under continuum approximation can be when the moiré potential is considered can be written down as,

$$\mathcal{H} = \mathcal{H}_0(\mathbf{k} \rightarrow -i\partial_{\mathbf{r}}) + V(\mathbf{r}), \quad (5.7)$$

where $\mathcal{H}_0$ is the original Hamiltonian and $V(\mathbf{r})$ is the real space moiré potential we discussed before. This Hamiltonian can be solved using a series of plane waves. Possible novel physical phenomena arising from this solution will be discussed in the following section.

5.3 Emergent band topology

It is natural to define a valley Chern number for bilayer graphene (BLG) under a superlattice potential, which makes the valley Hall effect possible. More interestingly, this superlattice can also flatten the bands, enhancing electron-electron interactions. We first show the single particle topological phase diagram for such a system in Fig. 5.3. The amplitude of this superlattice potential is set to 5 meV and the lattice constant is set to be 50 nm. We find the physics discussed here will not be affected if we slightly change these two parameters. The distribution of the Chern number and the corresponding bandwidth of the valence band and conduction band versus the displacement field D and the shape of the potential ϕ is plotted. The numerical results are consistent with a recent research [253].

We can clearly see the topological phase transition of the valence band and conduction band by tuning the displacement field in the upper panels of Fig. 5.3. Combined with the bandwidth plotted in the lower panel, we can find a region $-20, \text{meV} < D < 20, \text{meV}, \phi \approx 0$ where the bandwidth of the first conduction band is small and has a non-trivial Chern number. This suggests a parameter space where strong electron-electron interactions may lead to novel correlated phases due to the combination of narrow bandwidth and non-trivial topology.

We then pick a specific case to gain a deeper understanding. As shown in Fig. 5.4, the band structures of the superlattice are displayed in panel (a). Two flat bands appear near the Fermi level, highlighted in red. From the phase diagram, we can confirm that the conduction band is non-trivial and carries a Chern number $\mathcal{C} = 1$. The corresponding Berry curvature and quantum metric distributions are shown in panels (b) and (c), respectively. The Berry curvature distribution here is different from what we discussed

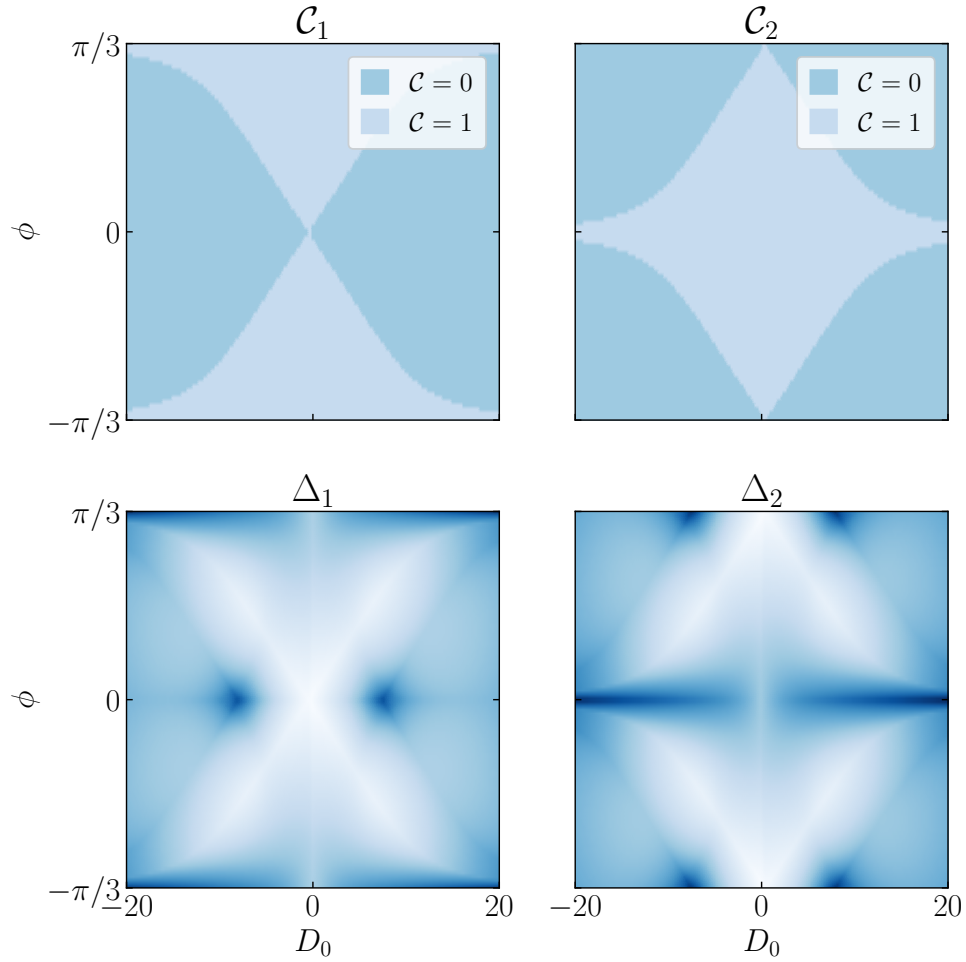


Figure 5.3: Single particle topological phase diagram for moiré BLG model. C_1 labels the valley ($\tau = 1$) Chern number for the valence band and C_2 labels the valley Chern number for the conduction band. Δ_1 labels the bandwidth of the valence band and Δ_2 labels the bandwidth of the conduction band. The dark area labels the region where the band width is small. The displacement field D is in the unit of meV and ϕ is in the unit of radius.

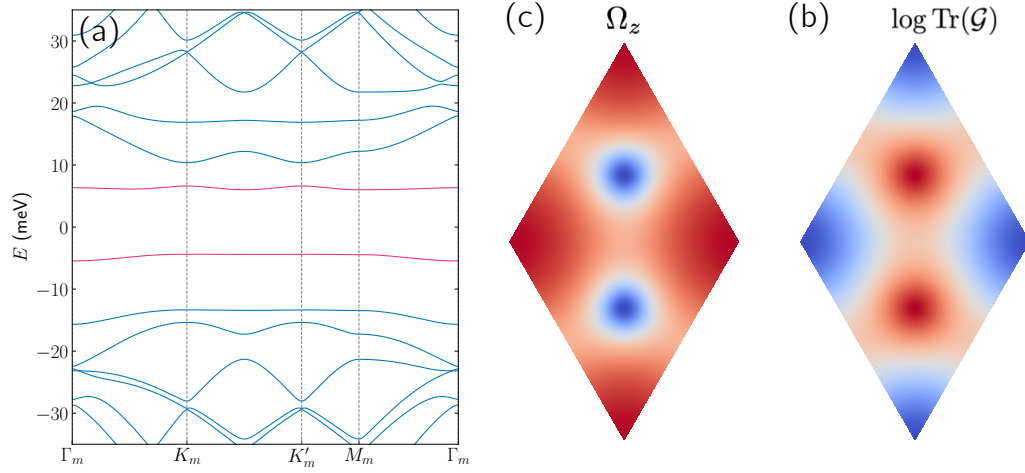


Figure 5.4: (a) The band structure of the BLG under a superlattice potential ($V_0 = 10$ meV, $\phi = 0$, $\tau = 1$). The conduction band valence band is labeled in red while other remote bands are in blue. We can clearly see these two bands are well separated from other bands. The conduction band carries $\mathcal{C} = 1$ and the valence band is topologically trivial. (b) The berry curvature (Ω_z) distribution in the reduced B.Z. The Γ_m point is located at the corner of the parallelogram. (c) The quantum metric distribution ($\log \text{Tr}(\mathcal{G})$) in the reduced B.Z.

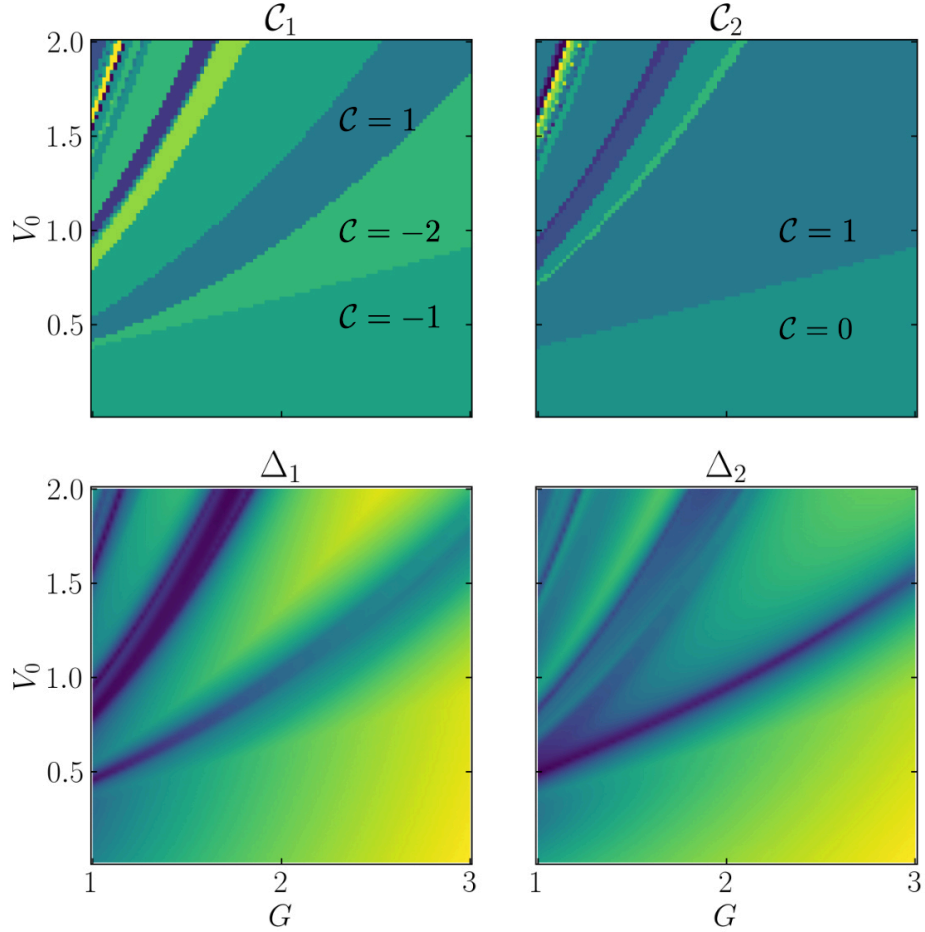


Figure 5.5: Single particle topological phase diagram for a simplified moiré BHZ model. $\mathcal{C}_1$ labels the spin Chern number for the valence band and $\mathcal{C}_2$ labels the spin Chern number for the conduction band. Δ_1 labels the bandwidth of the valence band and Δ_2 labels the bandwidth of the conduction band. The dark area highlights the region where the bandwidth is small, i.e. where flat bands appear.

in Chapter 4, where it was concentrated in regions where the gap is opened. In this case, the Berry curvature shows a relatively flat distribution, suggesting that the Fractional Quantum Hall Effect (FQHE) could potentially be engineered [243]. The origin of this topological behavior is discussed in detail in Ref. [253], which argues that holes in the valence bands tend to form a triangular lattice, while those in the conduction band tend to form a honeycomb lattice. This leads to the belief that a spin-valley polarized state, which is topologically non-trivial, can be achieved when the chemical potential is tuned to $\nu = 1$. This has also been confirmed through self-consistent Hartree-Fock calculations in Ref. [243, 253].

Next, we examine another case: the moiré BHZ model. We first simplify this model by neglecting the particle hole asymmetry ($C_1 = 0$), which was first studied in Ref. [254]. The single-particle phase diagram is depicted in Fig. 5.5, with the parameter space defined by varying the superlattice potential amplitude V_0 and the superlattice size L (where G represents the corresponding reciprocal lattice vector, and $L \approx 1/G$ represents the moiré lattice vector, other parameters δ, A, M_1 are all taken to be 1, we set $\phi = \pi$ in this calculation). For the moiré conduction band, we find a region where the bands are flat and possess a non-zero spin Chern number. Similarly, the valence bands can also be non-trivial with higher Chern numbers.

The band inversion in the BHZ model can be understood through a straightforward argument. When the band gap of the original band is large and the superlattice potential is weak, the bands do not invert, thus remaining trivial. However, when the original gap, denoted by δ , is small and comparable to the superlattice potential V_0 , band inversion can occur, driven by the moiré superlattice. This feature is illustrated in the right panel of Fig. 5.5, where a topological phase transition takes place as the potential amplitude increases.

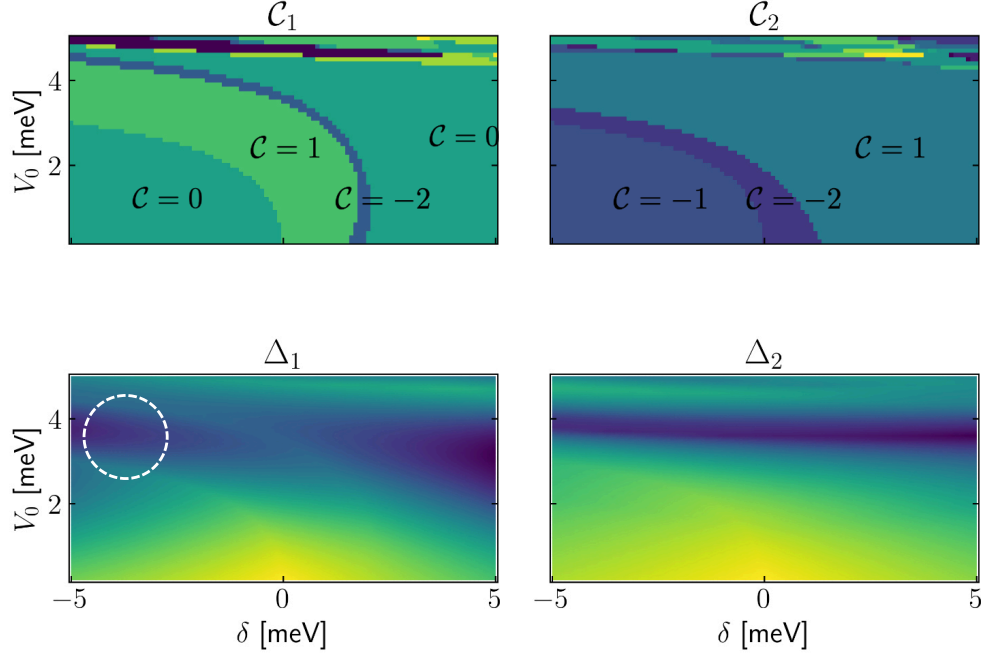


Figure 5.6: Single particle topological phase diagram for moiré Cd_3As_2 thin film under a C_{6z} symmetric moiré potential. C_1 labels the spin Chern number for the valence band and C_2 labels the spin Chern number for the conduction band. Δ_1 labels the bandwidth of the first valence band and Δ_2 labels first the bandwidth of the conduction band. A potential region to realize correlated Chern insulator is labeled in a dashed white circle. The dark area highlights the region where the bandwidth is small, i.e. where flat bands appear.

5.4 Correlated phases in the moiré BHZ model

In this section, we study a specific example of the moiré BHZ model, the (001) grown Cd_3As_2 thin film under a C_{6z} symmetric superlattice potential. As analyzed earlier, the (001) grown Cd_3As_2 thin films behave as a minimal BHZ model, which owns strong particle hole asymmetry. We now investigate possible physical effects when the thin film is engineered with a superlattice gate in the weak potential limit. Unlike the graphene system analyzed before, there is no degree of freedom for valley and spin, which provides this system with another advantage.

We first discuss the case when the potential has C_{6z} rotational symmetry ($\phi = 0$).

The full Hamiltonian then belongs to the D_{6h} point group. Due to the emergent $\mathcal{PT}$ symmetry, the band structures are doubly degenerate everywhere in momentum space, but we can still define the spin Chern number as discussed earlier.

Fig. 5.6 shows the single-particle phase diagram for a (001) grown Cd_3As_2 thin film, the moiré potential has a periodicity of 50 nm and this is assumed in the latter discussion, and consistent with the experimental setup [250]. The parameter δ controls the band gap of the unfolded band, indicating whether the original material is in the normal or inverted phase. V_0 represents the strength of the moiré potential. As shown in Fig. 5.6, there is a large region where the system's valence band is flat and carries a non-trivial spin Chern number.

We highlight a specific case ($\delta = -5$ meV, $V_0 = 3.5$ meV). The corresponding band structure is shown in Fig. 5.7-(a), where the Wilson loop diagram of the valence band suggests non-trivial topology. The bandwidth of this valence band is small, raising the question of whether enhanced Coulomb interaction will open a gap in such a system, especially when a hole per moiré B.Z. is doped.

Note that the strength of the electron electron interaction can be estimated through $e^2/4\pi\epsilon\epsilon_0|\mathbf{L}| \approx 3$ meV when $\epsilon = 10$, $|\mathbf{L}| = 50$ nm. This energy scale is comparable with the band width of the moiré bands in this scenario and suggests the effect from Coulomb interaction cannot be ignored. The interacting part of the Hamiltonian can be written in the plane wave basis,

$$H_{\text{int}} = \frac{1}{2N_{\mathbf{k}}} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} \sum_{\mu, \mu'} v_{\mathbf{q}} c_{\mathbf{k}+\mathbf{q}, \mu}^\dagger c_{\mathbf{k}', \mu'}^\dagger c_{\mathbf{k}', \mu'} c_{\mathbf{k}, \mu}, \quad (5.8)$$

where $\mu = (\tau, s)$ is the joint index for orbital and spin. Moreover, we adopt the double-

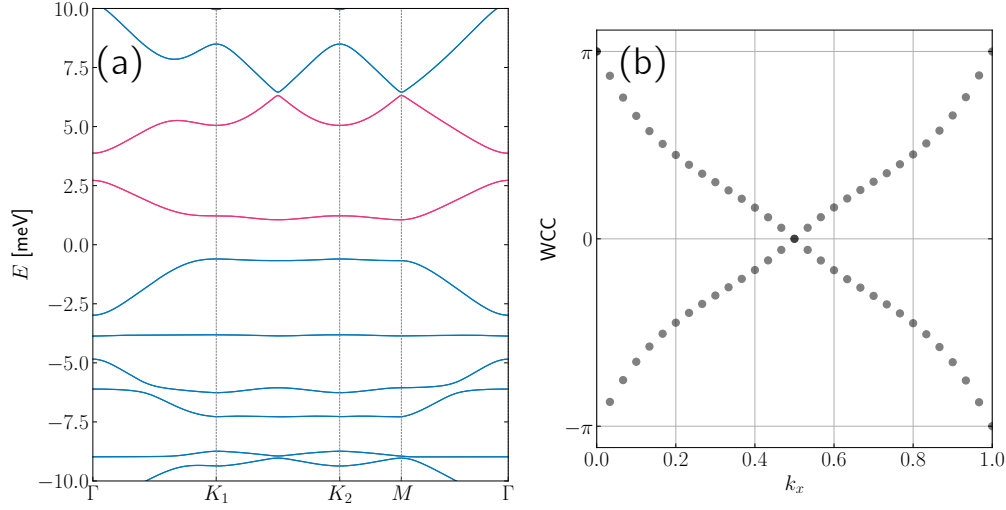


Figure 5.7: (a) moiré bands of the Cd_3As_2 under a C_{6z} symmetric potential with a periodicity of 50 nm. $V_0 = 3.5$ meV, $\delta = -5$ meV. The conduction bands and valence bands are highlighted in red and they are all doubly degenerate due to $\mathcal{PT}$ symmetry. (b) Wilson loop of the valence band. The winding feature hints the non-trivial topology.

gate screened interaction,

$$v_{\mathbf{q}} = \frac{e^2}{2\epsilon_0\epsilon_r\Omega_s} \frac{\tanh(d_s q)}{q}. \quad (5.9)$$

where $q = |\mathbf{q}|$, Ω_s is the size of the moiré unit cell and ϵ_0 is the permittivity of vacuum. We further fix the gate distance $d_s = 10$ nm in our later calculations. We project the full Hamiltonian into the active band subspace (two valence bands in this case) and perform momentum space unconstrained Hartree-Fock (UHF) calculations. We define the single particle density matrix $\rho_{mn}(\mathbf{k}) = \langle d_m^\dagger(\mathbf{k})d_n(\mathbf{k}) \rangle$ ¹ as the order parameter, where $d^\dagger(\mathbf{k})$, $d(\mathbf{k})$ are creation operator and annihilation operator defined in the projected band basis, respectively. The initial guess for $\rho(\mathbf{k})$ is constructed by projecting $\sigma_i \otimes \tau_j \otimes \mathbf{1}$ onto the active bands. The criterion for convergence of the self-consistent calculations is that the difference between the updated and previous density matrices

¹Another common definition for the single particle density matrix is $\rho_{nm}(\mathbf{k}) = \langle d_m^\dagger(\mathbf{k})d_n(\mathbf{k}) \rangle$, which is the transpose of the one defined in the main text.

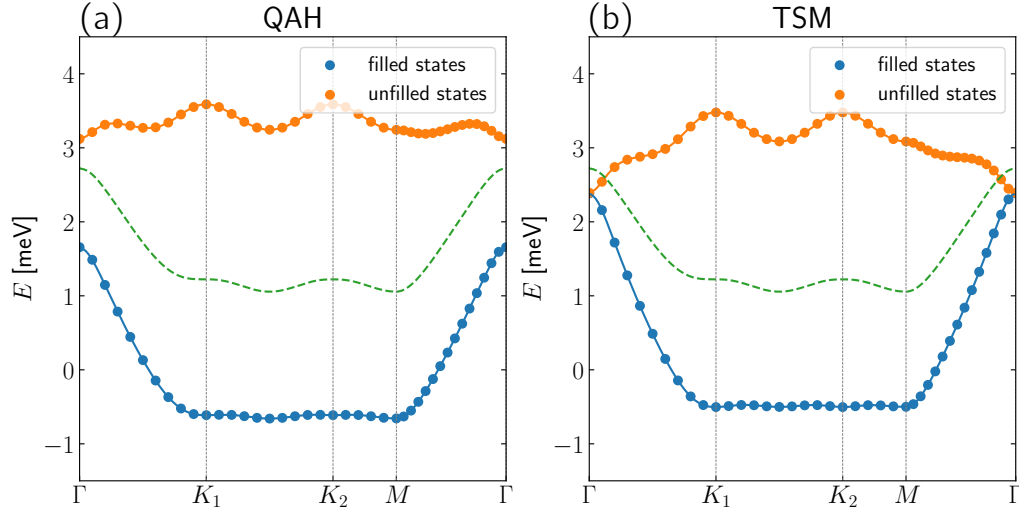


Figure 5.8: Hartree Fock mean-field solutions for the filling $\nu = -1$. The original bands are labeled in green dashed line and the interpolated quasi particle bands are labeled in the solid line. The blue dots represent the filled states while the orange dots label the unfilled states. (a) Quantum anomalous Hall (Chern insulator) phase (b) Topological semi-metal phase. Both phases can benefit from spontaneously symmetry breaking.

is less than 10^{-9} ($\sum_{\mathbf{k}} \|\rho_{i+1}(\mathbf{k}) - \rho_i(\mathbf{k})\| < 10^{-9}$). For more details of the band projected Hartree Fock methods, see Appendix. B.1.

We then perform HF calculations on a dense mesh with 30×30 $\mathbf{k}$ points, starting with the guessed density matrix. The results converged into two outcomes, shown in Fig. 5.8. The left panel depicts an insulating phase that spontaneously breaks time-reversal symmetry and $C_{2z}\mathcal{T}$ symmetry. More interestingly, the bands carry non-trivial Chern numbers of ± 1 , suggesting a quantum anomalous Hall insulator phase and this is confirmed by integrating out the berry curvature, shown in Fig. 5.9-(b). This phase owns a “spin-polarized” (SP) nature, which is confirmed by inspecting the component of the wavefunctions. The right panel depicts a semi-metal phase that breaks time-reversal symmetry and has a spin coherent feature while preserving $C_{2z}\mathcal{T}$ symmetry (this can be checked by the Berry curvature distribution in momentum space, which

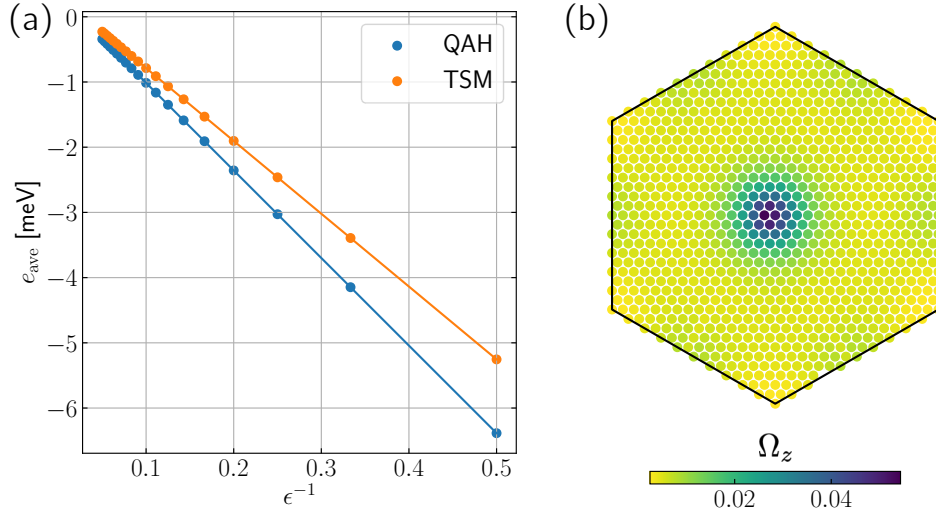


Figure 5.9: (a) Comparison of total energy per particle of the mean field solutions for the quantum anomalous Hall phase and topological semi-metal phase by varying the relative permittivity ϵ . (b) The calculated berry curvature (Ω_z) distribution in the moiré B.Z. of the first valence band in the quantum anomalous Hall phase.

shows singularities only at the band degenerate points and is zero elsewhere). The node at the Γ point cannot be gapped out by Coulomb interaction, and this kind of semi-metal phase is also found in the MATBG system. More symmetries consideration is listed in Table 5.1. These two interacting phases both benefit from energy reduction by spontaneously breaking symmetries when a hole is doped, which can be directly concluded from the mean field quasi-particle band structure. These two phases compete with each other when the Coulomb interaction is turned on and we further found that the insulating phase is more energy favorable if the interaction is strengthened, as shown in Fig. 5.9-(a). This is diagnosed by investigating the energy per particle,

$$\begin{aligned}
 e_{\text{ave}} &= \frac{1}{N_{\mathbf{k}}} \left(\sum_{\mathbf{k}} \text{Tr}[H_{\text{mf}}[\rho](\mathbf{k})\rho(\mathbf{k})^T] - E_c[\rho] \right) \\
 &= \frac{1}{N_{\mathbf{k}}} \left(\sum_{\mathbf{k}} \text{Tr}[H_0[\rho](\mathbf{k})\rho(\mathbf{k})^T] + \frac{1}{2} \sum_{\mathbf{k}} \text{Tr}[H_h[\rho](\mathbf{k})\rho(\mathbf{k})^T] - \frac{1}{2} \sum_{\mathbf{k}} \text{Tr}[H_f[\rho](\mathbf{k})\rho(\mathbf{k})^T] \right)
 \end{aligned} \tag{5.10}$$

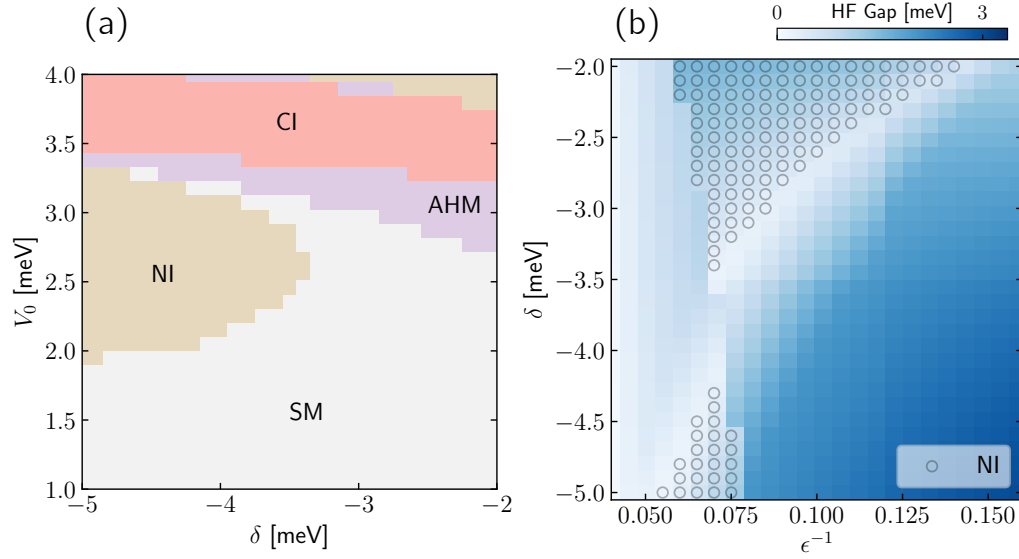


Figure 5.10: Mean field phase diagram for the Cd_3As_2 thin films under C_{6z} moiré potential (a) The original band gap δ and moiré potential strength are set as parameters and the relative permittivity ϵ is set as 25. The Chern insulator (CI) phase can be stabilized and anomalous Hall metal phase (AHM) also appears. SM denotes the semi-metal phase and NI denotes the normal insulator phase. (b) The original band gap δ and the relative permittivity ϵ are set as tuning parameters. The increasing of interacting strength will result in band mixing and topological phase transition happens.

where $E_c[\rho]$ is the condensation energy and the prefactor of $1/2$ is used to deduct the condensation energy. Note that, in the above calculations, we mainly focus on this two band model. Band mixing effect may also brings normal insulator phase if more bands are taken into consideration, making the problem more complicated. This will be discussed later using a six band model in the following discussion.

Previous discussions show the possibility to obtain a SP-QAH phase in a two band model. However, the bands we concerned about will also interact with the adjacent bands if we turn on the Coulomb interaction. Then, we will consider these effects in the following discussions. The full picture of the Hartree-Fock mean-field phase diagram is shown in Fig. 5.10. This diagram is calculated at zero temperature on a 15×15

k -mesh by projecting the full Hamiltonian onto six bands (four valence bands and two conduction bands) to fully account for band mixing effects when interactions are considered. We first fix $\epsilon = 25$, indicating that the interaction is at an intermediate level. The phase diagram is shown in panel (a). We found that the Chern insulator phase ($|C| = 1$) can be stabilized by tuning the band gap δ and the moiré potential strength V_0 . Furthermore, the anomalous Hall metal (AHM) phase appears, indicating no global band gap in the mB.Z. while the bands carry non-trivial Berry curvature. We then studied how interaction strength affects the Chern insulator phase in panel (b). Here, the moiré potential strength is fixed at 3.5 meV, and the relative permittivity ϵ is used as a tuning parameter. The normal insulator (NI) phase is labeled with white circles, while other areas in the phase diagram are in the CI phase. Increasing interaction strength initially causes band mixing, pushing the Chern insulator phase into a normal insulator phase. However, with further increase in interaction strength, the CI phase can be reentrant, and the HF band gap can be enhanced to around 3 meV. We also present a brief discussion when a C_{4z} symmetric potential is applied in the Appendix. [B.2](#).

Table 5.1: Symmetries of the Quantum anomalous Hall (QAH) phase and the topological semi-metal (TSM) phase

Phases/Symmetry	$\mathcal{T}$	$C_{2z}\mathcal{T}$	$\mathcal{M}_z$	$\mathcal{P}$	C_{2z}
QAH	×	×	✓	✓	✓
TSM	×	✓	×	✓	×

5.5 Conclusion

In this chapter, we studied the band topology of two artificial moiré lattices: one is Bernal bilayer graphene (BLG) and the other is the Bernevig-Hughes-Zhang (BHZ)

model under a superlattice potential. Unlike twisted bilayer materials, the moiré potential in these systems is generated by interlayer interactions and long-period mechanical modulations, such as lattice relaxation and strain. In the case we studied here, the material itself remains uniform and clean while being subjected to a real-space static electrical potential. This potential is generated by a patterned gate or a twisted substrate, creating a superlattice. By carefully designing the superlattice potential and tuning the system parameters, it is possible to achieve flat bands with non-zero Chern numbers, paving the way for realizing exotic quantum phenomena such as the quantum anomalous Hall effect and the Fractional Quantum Hall Effect. We use a concrete example, the (001) grown Cd_3As_2 under a C_{6z} symmetric moiré superlattice potential to show the Quantum Anomalous Hall effect can be available in the experimental feasible region under mean field theory. It will also be interesting to investigate whether such long period modulation can spontaneously appear when the system is slightly doped by forming Wigner crystal or anomalous Hall crystal [255–261]. The ability to control and manipulate these topological states in artificial moiré lattices opens up new avenues for research in condensed matter physics and materials science.

Chapter 6

Summary and outlook

In this dissertation, we delve into the quantum behavior of two representative low-dimensional materials from theoretical and computational perspectives: twisted bilayer graphene (TBG) and cadmium arsenide (Cd_3As_2) thin films.

Twisted Bilayer Graphene (TBG)

Twisted bilayer graphene is a prominent material in the moiré family, known for hosting many quantum phases in such system. To understand its low-energy behavior considering lattice relaxation and corrugation, we present a highly efficient and accurate numerical method in Chapter 2. This methodology named Truncated Atomic Plane Wave (TAPW) method, interestingly, can be combined with first-principle outputs to study twisted transition metal dichalcogenides (TMDs). This numerical method shows great potential in accelerating the prediction of twisted bilayer systems in a single-particle manner. A natural future direction is to integrate the TAPW method with first-principle computational packages like OpenMX and develop post-processing

codes to obtain topological properties and optical responses in a non-orthogonal basis.

In Chapter 3, we leverage this new method to study the moiré phonons and effective electron-phonon coupling in magic-angle twisted bilayer graphene. We demonstrate that the low-frequency phonons exhibit rich vibrational features, including patterns consistent with those observed in STM experiments. For optical phonons, we identify a specific branch originating from the transverse optical phonon of monolayer graphene that couples strongly with flat-band electrons, potentially opening a considerable gap if this vibrational mode is frozen.

Cadmium Arsenide (Cd_3As_2)

In Chapter 4, we explore the topological properties and transport features of cadmium arsenide thin films when subjected to a magnetic field. (001) grown Cd_3As_2 thin films are predicted to be 2D topological insulators when the film thickness is well-controlled. This is confirmed by Landau fan diagrams with inverted zeroth Landau levels. When the magnetic field is confined to the x - y plane, the Zeeman effect dominates, leading to the appearance of a 2D Weyl semi-metal phase, which is confirmed both theoretically and experimentally. We further predict that if the growth orientation is tuned to the (112) direction, the quantized in-plane anomalous Hall effect can manifest when the film is near the critical thickness due to the geometry obstruction.

The experimentally confirmed 2D topological insulator phase of Cd_3As_2 serves as an effective Bernevig-Hughes-Zhang (BHZ) model. We then study how an effective superlattice potential affects the band topology of the material in Chapter 5. It is predicted that a spin-polarized state can emerge as the ground state if the parameters in the BHZ model are well controlled, hosting a non-trivial Chern number due to moiré-

enhanced electron-electron interactions. A significant advantage of this system is the well-controlled strain, which is not easily achievable in other 2D materials. However, challenges remain, particularly in estimating the effective moiré potential in superlattice gating, which is also future subject of study.

Appendix A

Appendix for Truncated Atomic Plane Wave method

A.1 Symmetry and boundary conditions

In this section, we will briefly discuss mathematical properties of TAPW basis under symmetry operations. In the continuous limit, the TAPW basis can be approximated as plane waves,

$$\begin{aligned} |\psi_{\alpha n}(\bar{\mathbf{k}})\rangle &\approx \frac{1}{\sqrt{N_m N_a}} \sum_{\mathbf{I}_i} e^{i(\bar{\mathbf{k}} + \mathbf{G}_n) \cdot \mathbf{r}} |\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha}\rangle \\ &= \frac{e^{i(\bar{\mathbf{k}} + \mathbf{G}_n) \cdot \mathbf{r}}}{\sqrt{N_m \Omega}} \left(\sqrt{\frac{\Omega}{N_a}} \sum_{\mathbf{I}_i} |\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha}\rangle \right) \end{aligned} \quad (\text{A.1})$$

It is clear that the plane wave envelopes the atomic details in the bracket. Then, the atomic plane wave basis is actually the plane wave we are familiar with when we treat the problem of free electrons. From now on we denote our basis set as (now $\alpha \rightarrow (\alpha; \ell)$),

α is sublattice index and ℓ is the layer index.)

$$|\bar{\mathbf{k}} + \mathbf{G}, \alpha \ell \eta s\rangle = \frac{1}{\sqrt{N_m N_a}} \sum_{\mathbf{I}i} e^{i(\bar{\mathbf{k}} + \mathbf{G}_n) \cdot (\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})} \phi(\mathbf{r} - (\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})), \quad (\text{A.2})$$

In the above equation, η, s are indices for valley and spin, respectively.

A.1.1 Discrete symmetry for TAPW basis

Let's focus on several important discrete symmetries in TBG system. They are C_{3z} , C_{2x} , C_{2z} and time reversal $\mathcal{T}$.

For C_{3z} ,

$$\begin{aligned} C_{3z} |\bar{\mathbf{k}} + \mathbf{G}, \alpha \ell \eta s\rangle &= \frac{1}{\sqrt{N_m N_a}} \sum_{\mathbf{I}i} e^{iC_{3z}(\bar{\mathbf{k}} + \mathbf{G}_n) \cdot C_{3z}(\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})} \phi(C_{3z}^{-1} \mathbf{r} - (\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})) \\ &= \frac{1}{\sqrt{N_m N_a}} \sum_{\mathbf{I}i} e^{iC_{3z}(\bar{\mathbf{k}} + \mathbf{G}_n) \cdot C_{3z}(\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})} \phi(\mathbf{r} - C_{3z}(\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})) \quad (\text{A.3}) \\ &= \frac{1}{\sqrt{N_m N_a}} \sum_{\mathbf{I}i} e^{iC_{3z}(\bar{\mathbf{k}} + \mathbf{G}_n) \cdot (\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})} \phi(\mathbf{r} - (\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})) \end{aligned}$$

Note that, $C_{3z}(\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})$ will recover the sublattice and $C_{3z}(\bar{\mathbf{k}} + \mathbf{G}_0 + \mathbf{G}_n) = C_{3z}\bar{\mathbf{k}} + C_{3z}\mathbf{G}_n + \mathbf{G}_0 - \eta\mathbf{b}_2$, then we have:

$$C_{3z} |\bar{\mathbf{k}} + \mathbf{G}, \alpha \ell \eta s\rangle = e^{-i\mathbf{b}_2 \cdot \boldsymbol{\tau}_\alpha} |C_{3z}\bar{\mathbf{k}} + C_{3z}\mathbf{G}, \alpha \ell \eta s\rangle. \quad (\text{A.4})$$

For C_{2z} , $C_{2z}(\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})$ will exchange the sublattice and $C_{2z}(\bar{\mathbf{k}} + \mathbf{G}_0 + \mathbf{G}_n) = -\bar{\mathbf{k}} - \mathbf{G}_0 - \mathbf{G}_n$. These analysis is exactly true for a D_6 structure but approximate for a D_3 structure.

$$C_{2z} |\bar{\mathbf{k}} + \mathbf{G}, \alpha \ell \eta s\rangle = |-\bar{\mathbf{k}} - \mathbf{G}, \bar{\alpha} \ell \bar{\eta} s\rangle \quad (\text{A.5})$$

For C_{2x} , $C_{2x}(\mathbf{R}_I + \boldsymbol{\tau}_{i\alpha\ell})$ will exchange the sublattice and layer, it requires,

$$C_{2x} |\bar{\mathbf{k}} + \mathbf{G}, \alpha\ell\eta s\rangle = |C_{2x}\bar{\mathbf{k}} + C_{2x}\mathbf{G}, \bar{\alpha}\bar{\ell}\eta s\rangle. \quad (\text{A.6})$$

For spinless time reversal symmetry (TRS) $\mathcal{T}$,

$$\mathcal{T} |\bar{\mathbf{k}} + \mathbf{G}, \alpha\ell\eta s\rangle = |-\bar{\mathbf{k}} - \mathbf{G}, \alpha\ell\bar{\eta} s\rangle. \quad (\text{A.7})$$

Note that TRS is anti-unitary, which can be written as $T = UK$ where U is the unitary part of the operator and K represents for the complex conjugate. For an arbitrary operator O , $TOT^{-1} = UKOKU^\dagger = UO^*U^\dagger$. This property also holds for symmetry operator like $C_{2z}T$. For TBG, TRS further requires:

$$H_{\mathbf{G}\alpha\ell, \mathbf{G}'\beta\ell'}^\eta(\bar{\mathbf{k}}) = [H_{-\mathbf{G}\alpha\ell, -\mathbf{G}'\beta\ell'}^{\bar{\eta}}(-\bar{\mathbf{k}})]^*. \quad (\text{A.8})$$

Based on above analysis, we can directly write down the corresponding creation operators under symmetry operations:

$$\begin{aligned} C_{3z} c_{\bar{\mathbf{k}}, \mathbf{G}, \alpha, \ell, \eta, s}^\dagger C_{3z}^{-1} &= [e^{i\frac{2\pi}{3}\sigma_z\tau_z}]_{\alpha'\eta', \alpha\eta} c_{C_{3z}\bar{\mathbf{k}}, C_{3z}\mathbf{G}, \alpha, \ell, \eta, s}^\dagger, \\ (C_{2z}T) c_{\bar{\mathbf{k}}, \mathbf{G}, \alpha, \ell, \eta, s}^\dagger (C_{2z}T)^{-1} &= c_{\bar{\mathbf{k}}, \mathbf{G}, \bar{\alpha}, \bar{\ell}, \eta, s}^\dagger, \\ (C_{2x}) c_{\bar{\mathbf{k}}, \mathbf{G}, \alpha, \ell, \eta, s}^\dagger (C_{2x})^{-1} &= c_{C_{2x}\bar{\mathbf{k}}, C_{2x}\mathbf{G}, \bar{\alpha}, \bar{\ell}, \eta, s}^\dagger. \end{aligned} \quad (\text{A.9})$$

In the continuous limit, the matrix representation for the symmetry operators are:

$$D(C_{2z}T) = \tau_0 \sigma_x \varrho_0, D(C_{2x}) = \tau_0 \sigma_x \varrho_x, D(C_{3z}) = e^{i\frac{2\pi}{3}\sigma_z\tau_z} \varrho_0. \quad (\text{A.10})$$

where $\sigma_{x,y,z}(\sigma_0)$, $\tau_{x,y,z}(\tau_0)$, $\varrho_{x,y,z}(\varrho_0)$ are pauli matrices (identity matrix) for sublattice,

valley, layer degree of freedom.

A.1.2 PBC in TAPW basis

As stated in the last section, TAPW basis is plane wave under continuum approximation and envelope function approximation. After solving the Hamiltonian, the eigen wavefunction is indeed Bloch function:

$$\psi_{\bar{\mathbf{k}}} = \sum_{\mathbf{G}_j} C_{\mathbf{G}_j}(\bar{\mathbf{k}}) e^{i(\bar{\mathbf{k}} + \mathbf{G}_j) \cdot \mathbf{r}}. \quad (\text{A.11})$$

In addition, it should satisfy

$$\psi_{\bar{\mathbf{k}} + \mathbf{G}} = \psi_{\bar{\mathbf{k}}} = \sum_{\mathbf{G}_i} C_{\mathbf{G}_i}(\bar{\mathbf{k}} + \mathbf{G}) e^{i(\bar{\mathbf{k}} + \mathbf{G} + \mathbf{G}_i) \cdot \mathbf{r}} \quad (\text{A.12})$$

which leads to

$$C_{\mathbf{G}_i}(\bar{\mathbf{k}} + \mathbf{G}) = \delta_{\mathbf{G}_i + \mathbf{G}, \mathbf{G}_j} C_{\mathbf{G}_j}(\bar{\mathbf{k}}) \quad (\text{A.13})$$

The corresponding matrix form is

$$\mathbf{D}(\bar{\mathbf{k}} + \mathbf{G}) = \mathbf{T}(-\mathbf{G}) \mathbf{D}(\bar{\mathbf{k}}) \quad (\text{A.14})$$

where $(\mathbf{T}(\mathbf{G}))_{ij} = \delta_{\mathbf{G}_i, \mathbf{G}_j + \mathbf{G}}$. Note that: $\mathbf{T}(-\mathbf{G}) = \mathbf{T}(\mathbf{G})^T$. In the above derivation, we omit the indices for sublattice. PBC is important when performing Wilson loop calculation to determine the topology of the Bloch bands. Another quantity we will focus on is:

$$C_{\mathbf{G}_i + \mathbf{G}, n}(\bar{\mathbf{k}}) = \delta_{\mathbf{G}_i + \mathbf{G}, \mathbf{G}_j} C_{\mathbf{G}_j, n}(\bar{\mathbf{k}}) \quad (\text{A.15})$$

where n is a band index.

Table A.1: Six largest components of moiré potential $U_{\alpha\mathbf{G}_0,\beta\mathbf{G}_i}(\bar{\mathbf{k}})$ on high symmetry points when $\theta = 1.085^\circ$ and a corrugated structure as an input. The unit is electron Voltage (eV). Unlike classical BM model which sets $u \neq u'$, our method has a detailed description for higher orders components of moiré potential.

$U_{A_1A_2}(\bar{\Gamma})$	0.085809	0.079906	0.074568	0.010323	0.010078	0.009875
$U_{A_1B_2}(\bar{\Gamma})$	0.103212	0.097788	0.092846	0.007722	0.007437	0.007261
$U_{B_1A_2}(\bar{\Gamma})$	0.103212	0.097788	0.092846	0.007722	0.007437	0.007261
$U_{B_1B_2}(\bar{\Gamma})$	0.085809	0.079906	0.074568	0.010323	0.010078	0.009874
$U_{A_1A_2}(\bar{K})$	0.080045	0.080045	0.080045	0.010090	0.010090	0.010090
$U_{A_1B_2}(\bar{K})$	0.097890	0.097890	0.097890	0.007460	0.007460	0.007460
$U_{B_1A_2}(\bar{K})$	0.097890	0.097890	0.097890	0.007460	0.007460	0.007460
$U_{B_1B_2}(\bar{K})$	0.080045	0.080045	0.080045	0.010089	0.010089	0.010089
$U_{A_1A_2}(\bar{K}')$	0.085737	0.080045	0.074502	0.010300	0.010123	0.009853
$U_{A_1B_2}(\bar{K}')$	0.103157	0.097890	0.092799	0.007676	0.007527	0.007217
$U_{B_1A_2}(\bar{K}')$	0.103157	0.097890	0.092799	0.007676	0.007527	0.007217
$U_{B_1B_2}(\bar{K}')$	0.085737	0.080045	0.074502	0.010300	0.010123	0.009853
$U_{A_1A_2}(\bar{M})$	0.082864	0.080062	0.077246	0.010195	0.010103	0.009972
$U_{A_1B_2}(\bar{M})$	0.100496	0.097903	0.095316	0.007565	0.007488	0.007336
$U_{B_1A_2}(\bar{M})$	0.100496	0.097903	0.095316	0.007565	0.007488	0.007336
$U_{B_1B_2}(\bar{M})$	0.082864	0.080062	0.077246	0.010195	0.010103	0.009971

A.2 Higher order Fourier components of moiré potential

We tabulate the six largest Fourier components of the moiré potential $U(\mathbf{r})$ for MATBG ($\theta = 1.085^\circ$) at high symmetry $\bar{\mathbf{k}}$ points using our TAPW method with Slater Koster TB parameters in Table. A.1. TAPW method can help extract effective parameters in generalized BM model by reading these Fourier components.

A.3 Details on the force field

The potential energy for TBG can be expressed as a summation of the bonded intralayer interactions and non-bonded interlayer interactions.

$$U = U_{\text{bonded}} + U_{\text{non-bonded}}, \quad (\text{A.16})$$

where the bonded interactions can be modelled as $U_{\text{bonded}} = U_2 + U_3 + U_4$ by the Dreiding potential [180], which includes the bond stretch U_2 (two-body term), angle bend U_3 (three-body term), and dihedral torsion U_4 (four-body term).

Specifically, the bond stretch interactions between carbon i and carbon j can be described by a simple harmonic oscillator as $U_2^{ij} = \frac{1}{2}k_{ij}(R - R_0)^2$, where R_0 is the equilibrium bond length (i.e. 1.42 Å for graphene), and k_{ij} is a constant set as 700 kcal/(mol · Å²). The three-body angle bend formed by two bonds of atoms ij and jk which share a common atom j can be expressed in a harmonic cosine form as $U_3^{ijk} = \frac{1}{2}C_{ijk}(\cos \theta_{ijk} - \cos \theta_j^0)^2$. In the equation, θ_{ijk} is the angle between bonds ij and jk , and $\theta_j^0 = 120^\circ$ is an equilibrium angle. C_{ijk} is a constant set as 133.33 kcal/(mol · rad²). The four-body dihedral interactions $ijkl$ which consists of two bonds ij and kl connected via a common bond jk can be described by following form as: $U_4^{ijkl} = \frac{1}{2}V_{jk}\{1 - \cos[n_{jk}(\varphi - \varphi_{jk}^0)]\}$, where φ is the dihedral angle between ijk and jkl planes and $\varphi_{jk}^0 = 180^\circ$ is the equilibrium dihedral angle. n_{jk} is the periodicity, which is set as 2, and V_{jk} is the barrier to rotation, which is set as 5 kcal/mol.

The non-bonded van der Waals interactions $U_{\text{non-bonded}}$ between interlayer TBG can be expressed by the exponential-6 (X6) form [181] as $U_{\text{vdW}}^{\text{X6}} = Ae^{-R/c} - BR^{-6}$, where R is the distance between two interlayer atoms. The parameters A , c and B are set as 385631.5 kcal/mol, 0.2343 Å and 303.82 kcal/(mol·Å⁶). The X6 form has more

accurate description regarding the short-range interactions [\[181\]](#).

Appendix B

Appendix for momentum space Hartree Fock method

B.1 Band projected Hartree Fock method

In this appendix, we will briefly review the momentum space Hartree Fock method used in the main text. The interacting part of the Hamiltonian written in the plane wave basis is,

$$H_{\text{int}} = \frac{1}{2S} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} \sum_{\mu\mu'} v(\mathbf{q}) c_{\mathbf{k}+\mathbf{q},\mu}^\dagger c_{\mathbf{k}'-\mathbf{q},\mu'}^\dagger c_{\mathbf{k}',\mu'} c_{\mathbf{k},\mu}, \quad (\text{B.1})$$

where μ, μ' are joint indices for orbital, spin, valley, etc. and S is the total area. By considering the moiré structure or density wave orders,

$$H_{\text{int}} = \frac{1}{2S} \sum_{\bar{\mathbf{k}}\bar{\mathbf{k}}'\bar{\mathbf{q}}} \sum_{\mathbf{G}\mathbf{G}'\mathbf{Q}} \sum_{\mu\mu'} v(\bar{\mathbf{q}} + \mathbf{Q}) c_{\mathbf{G}+\mathbf{Q},\mu}^\dagger(\bar{\mathbf{k}} + \bar{\mathbf{q}}) c_{\mathbf{G}'-\mathbf{Q},\mu'}^\dagger(\bar{\mathbf{k}}' - \bar{\mathbf{q}}) c_{\mathbf{G}',\mu'}(\mathbf{k}') c_{\mathbf{G},\mu}(\mathbf{k}). \quad (\text{B.2})$$

For simplicity, we denote $\mathbf{G} = (\mathbf{G}, \mu)$ for simplicity. Based on this Hamiltonian, we can perform the band projected Hartree Fock calculation to save the computational power. For the band projected Hartree Fock Hamiltonian, we note that,

$$\begin{aligned} c_{\mathbf{G}} &= \sum_n D_{\mathbf{G}n} d_n, \\ c_{\mathbf{G}}^\dagger &= \sum_n D_{\mathbf{G}n}^* d_n^\dagger, \end{aligned} \quad (\text{B.3})$$

and $d_n^\dagger$ is the creation operator for band n and $D_{\mathbf{G}n}$ denotes the corresponding eigen wavefunction. The projected interacting Hamiltonian now should be,

$$\begin{aligned} H_{\text{int}} &= \frac{1}{2S} \sum_{\bar{\mathbf{k}}\bar{\mathbf{k}}'\bar{\mathbf{q}}} \sum_{\mathbf{G}\mathbf{G}'\mathbf{Q}} \sum_{mnm'n'} \sum_{\mu\mu'} v(\bar{\mathbf{q}} + \mathbf{Q}) D_{\mathbf{G}+\mathbf{Q},m}^* (\bar{\mathbf{k}} + \bar{\mathbf{q}}) D_{\mathbf{G}'-\mathbf{Q},m'}^* (\bar{\mathbf{k}}' - \bar{\mathbf{q}}) D_{\mathbf{G}',n'} (\bar{\mathbf{k}}') D_{\mathbf{G},n} (\bar{\mathbf{k}}) \\ &\quad d_m^\dagger (\bar{\mathbf{k}} + \bar{\mathbf{q}}) d_{m'}^\dagger (\bar{\mathbf{k}}' - \bar{\mathbf{q}}) d_{n'} (\bar{\mathbf{k}}') d_n (\bar{\mathbf{k}}). \end{aligned} \quad (\text{B.4})$$

Then we can define the form factor,

$$\lambda_{mn}(\mathbf{k}, \mathbf{q}, \mathbf{Q}) = \sum_{\mathbf{G}} D_{\mathbf{G}+\mathbf{Q},m}^* (\bar{\mathbf{k}} + \bar{\mathbf{q}}) D_{\mathbf{G},n} (\bar{\mathbf{k}}), \quad (\text{B.5})$$

such that, the interacting Hamiltonian can be written in a more compact way,

$$\begin{aligned} H_{\text{int}} &= \frac{1}{2S} \sum_{\bar{\mathbf{k}}\bar{\mathbf{k}}'\bar{\mathbf{q}}} \sum_{\mathbf{Q}} \sum_{mnm'n'} v(\bar{\mathbf{q}} + \mathbf{Q}) \lambda_{mn}(\bar{\mathbf{k}}, \bar{\mathbf{q}}, \mathbf{Q}) \lambda_{m'n'}(\bar{\mathbf{k}}', -\bar{\mathbf{q}}, -\mathbf{Q}) \\ &\quad d_m^\dagger (\bar{\mathbf{k}} + \bar{\mathbf{q}}) d_{m'}^\dagger (\bar{\mathbf{k}}' - \bar{\mathbf{q}}) d_{n'} (\bar{\mathbf{k}}') d_n (\bar{\mathbf{k}}). \end{aligned} \quad (\text{B.6})$$

where we can also define the Coulomb interaction tensor under the band basis,

$$U_{mnm'n'}(\bar{\mathbf{k}}, \bar{\mathbf{k}}', \bar{\mathbf{q}}) = \sum_{\mathbf{Q}} v(\bar{\mathbf{q}} + \mathbf{Q}) \lambda_{mn}(\bar{\mathbf{k}}, \bar{\mathbf{q}}, \mathbf{Q}) \lambda_{m'n'}(\bar{\mathbf{k}}', -\bar{\mathbf{q}}, -\mathbf{Q}) \quad (\text{B.7})$$

The Hartree interacting tensor now reads,

$$H_{mnm'n'}(\bar{\mathbf{k}}, \bar{\mathbf{k}}') = U_{mnm'n'}(\bar{\mathbf{k}}, \bar{\mathbf{k}}', 0) \quad (\text{B.8})$$

It is easy to check $H_{mnm'n'}(\mathbf{k}, \mathbf{k}') = H_{m'n'mn}(\bar{\mathbf{k}}', \bar{\mathbf{k}})$ because $v(\mathbf{Q})$ is $\mathbf{Q}$ rotationally symmetric. Furthermore, in the real calculation,

$$\begin{aligned} \lambda_{mn}(\bar{\mathbf{k}}, 0, \mathbf{Q}) &= \sum_{\mathbf{G}} D_{\mathbf{G}+\mathbf{Q},m}^*(\bar{\mathbf{k}}) D_{\mathbf{G},n}(\bar{\mathbf{k}}) \\ &= \sum_{ij} D_{\mathbf{G}_i,m}^*(\bar{\mathbf{k}}) D_{\mathbf{G}_j,n}(\bar{\mathbf{k}}) \delta_{\mathbf{G}_i, \mathbf{G}_j+\mathbf{Q}} \end{aligned} \quad (\text{B.9})$$

Note that,

$$\begin{aligned} \mathbf{T}(\mathbf{Q})_{ij} &= \delta_{\mathbf{G}_i, \mathbf{G}_j+\mathbf{Q}} \\ \mathbf{T}^T(\mathbf{Q})_{ij} &= \delta_{\mathbf{G}_i, \mathbf{G}_j-\mathbf{Q}} = \mathbf{T}(-\mathbf{Q}) \end{aligned} \quad (\text{B.10})$$

In our code, $\mathbf{T}(\mathbf{Q})$ is stored as `ttrans`, and

$$\begin{aligned} \lambda_{m'n'}(\bar{\mathbf{k}}', 0, -\mathbf{Q}) &= \sum_{\mathbf{G}} D_{\mathbf{G}-\mathbf{Q},m'}^*(\bar{\mathbf{k}}') D_{\mathbf{G},n'}(\bar{\mathbf{k}}') \\ &= \sum_{ij} D_{\mathbf{G}_i,m'}^*(\bar{\mathbf{k}}') D_{\mathbf{G}_j,n'}(\bar{\mathbf{k}}') \delta_{\mathbf{G}_i, \mathbf{G}_j-\mathbf{Q}}. \end{aligned} \quad (\text{B.11})$$

The Fock interacting tensor can be derived in the same way,

$$F_{m'nmn'}(\bar{\mathbf{k}}, \bar{\mathbf{k}}') = \sum_{\mathbf{Q}} v(\bar{\mathbf{k}}' - \bar{\mathbf{k}} + \mathbf{Q}) \left(\sum_{\mathbf{G}} D_{\mathbf{G}+\mathbf{Q},m}^*(\bar{\mathbf{k}}') D_{\mathbf{G},n}(\bar{\mathbf{k}}) \right) \left(\sum_{\mathbf{G}} D_{\mathbf{G}-\mathbf{Q},m'}^*(\bar{\mathbf{k}}) D_{\mathbf{G},n'}(\bar{\mathbf{k}}') \right). \quad (\text{B.12})$$

Note that, the orders of indices in the Fock term have been changed due to normal ordering. As a result, the total interacting tensor is defined as,

$$V(mn\bar{\mathbf{k}}, m'n'\bar{\mathbf{k}}') = H(mn\bar{\mathbf{k}}, m'n'\bar{\mathbf{k}}') - F(m'n\bar{\mathbf{k}}, mn'\bar{\mathbf{k}}'). \quad (\text{B.13})$$

The Hartree Fock correction for the original Hamiltonian should be,

$$H_{\text{hf}} = \frac{1}{N_{\bar{\mathbf{k}}}} \sum_{mn\bar{\mathbf{k}}} \sum_{m'n'\bar{\mathbf{k}}'} V_{mnm'n'}(\bar{\mathbf{k}}, \bar{\mathbf{k}}') \rho_{m'n'}(\bar{\mathbf{k}}') d_m^\dagger(\bar{\mathbf{k}}) d_n(\bar{\mathbf{k}}), \quad (\text{B.14})$$

such that the total Hamiltonian is corrected as $H_0 + H_{\text{hf}} - E_c$ and E_c is the condensation energy. The density matrix defined as following is the order parameter,

$$\rho_{m'n'}(\bar{\mathbf{k}}') = \left\langle d_{m'}^\dagger(\bar{\mathbf{k}}') d_{n'}(\bar{\mathbf{k}}') \right\rangle. \quad (\text{B.15})$$

which can be solved self-consistently. The total energy per particle is,

$$e = \frac{1}{N_{\bar{\mathbf{k}}}} \sum_{\bar{\mathbf{k}}} \left(\text{Tr}[H_0 \rho^T] + \frac{1}{2} \text{Tr}[H_{\text{hf}} \rho^T] \right), \quad (\text{B.16})$$

where the condensation energy is also deducted by the prefactor 1/2. The energy functional of the system is written as,

$$E[\rho] = \sum_{\bar{\mathbf{k}}mn} h_{mn}^0(\bar{\mathbf{k}}) \rho_{mn}(\bar{\mathbf{k}}) + \frac{1}{2N_{\bar{\mathbf{k}}}} \sum_{mn\bar{\mathbf{k}}} \sum_{m'n'\bar{\mathbf{k}}'} \rho_{mn}(\bar{\mathbf{k}}) V_{mnm'n'}(\bar{\mathbf{k}}, \bar{\mathbf{k}}') \rho_{m'n'}(\bar{\mathbf{k}}'). \quad (\text{B.17})$$

The second part of Eq. B.17 denoted as $E_{\text{int}}[\rho]$ is the interaction part of the energy functional. After expanding the density matrix using the generalized Pauli matrices Γ_{mn}^γ . We can define the Hartree Fock kernel,

$$K_{\gamma\mathbf{k},\gamma'\mathbf{k}'} = \frac{1}{2N_{\bar{\mathbf{k}}}} \sum_{mnm'n'} \Gamma_{mn}^\gamma V_{mnm'n'}(\bar{\mathbf{k}}, \bar{\mathbf{k}}') \Gamma_{m'n'}^{\gamma'}. \quad (\text{B.18})$$

This kernel is real and symmetric. It is natural to perform spectrum analysis and find the corresponding leading modes,

$$\begin{aligned} K_{\gamma\mathbf{k},\gamma'\mathbf{k}'} &= \sum_{\ell} K_{\ell} C_{\gamma\mathbf{k},\ell} C_{\ell,\gamma'\mathbf{k}'} \\ &\approx \sum_i K_i C_{\gamma\mathbf{k},i} C_{i,\gamma'\mathbf{k}'} \end{aligned} \tag{B.19}$$

We denote $C_{\gamma\mathbf{k},\ell}$ as the ℓ -th eigenvector of $K_{\gamma\mathbf{k},\gamma'\mathbf{k}'}$, with K_{ℓ} being the corresponding eigenvalue. From now on, we refer to $C_{\gamma\mathbf{k},\ell}$ as the ℓ -th eigenmode (or eigenchannel) of the Hartree-Fock kernel. Typically, several modes serve as the principal components of this kernel, suggesting that we only need to consider these modes rather than preserving all modes, as in the original band-projected Hartree-Fock procedure. In the second step of Eq. B.19, i represents the index for the principal components, and the ratio between $\sum_i K_i$ and $\sum_{\ell} K_{\ell}$ can be the criterion to determine whether this principal component analysis (PCA) is effective in a specific case.

B.2 C_{4z} symmetric case

In this section, we show present the two band Hartree Fock result and the six band phase diagram when a C_{4z} symmetric potential is applied in Fig. B.1. For the moiré potential, only $\mathbf{G}_1 = \frac{2\pi}{L}(1, 0)$, $\mathbf{G}_2 = \frac{2\pi}{L}(0, 1)$, $-\mathbf{G}_1$ and $-\mathbf{G}_2$ are preserved. Different from the case of C_{6z} symmetric potential, the Chern insulator phase cannot be stabilized by $\epsilon = 25$ and the Anomalous Hall metal (AHM) phase appear, which indicates the bands can obtain non-trivial berry curvature while there is no global band gap.

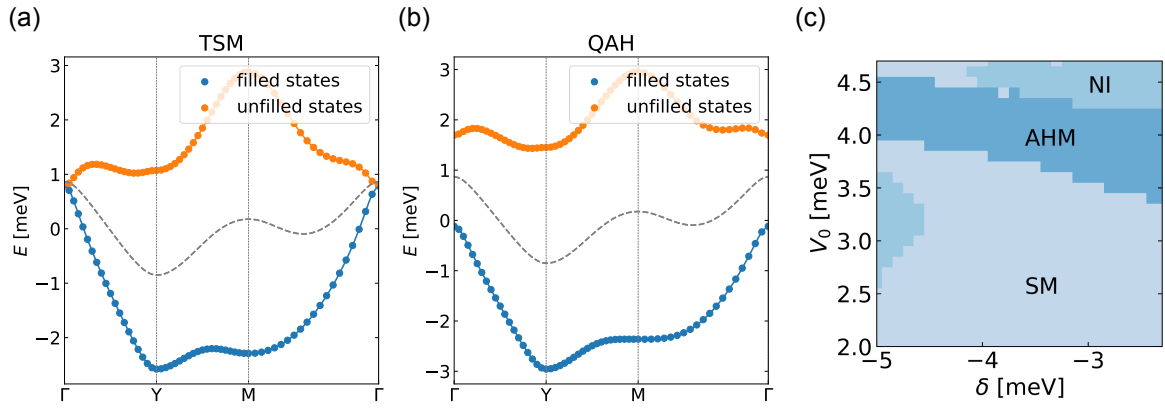


Figure B.1: Two band Hartree Fock calculations when a C_{4z} symmetric potential ($\delta = -5$ meV, $V_0 = 4$ meV) is applied. The self-consistent Hartree Fock calculations are performed on a 40×40 k -mesh. (a) Hartree Fock band structures for the spin-coherent state, exhibiting a topological semimetal (TSM) phase. The original bands are labeled in grey dashed line. (b) Hartree Fock band structures for the spin-polarized state, indicative of a quantum anomalous Hall (QAH) phase. (c) Mean field phase diagram for the Cd_3As_2 thin films under superlattice potential. The original band gap δ and moiré potential strength are set as parameters and the relative permittivity ϵ is set as 25 when a C_{4z} symmetric potential applied. The Chern insulator (CI) phase can not be stabilized but the anomalous Hall metal phase (AHM) appears. SM denotes the semi-metal phase and NI denotes the normal insulator phase.

B.3 Hartree Fock kernel analysis

We begin by discussing the potential order parameters stabilized by Coulomb interactions, analyzed through the eigenmodes of the Hartree-Fock interaction kernel. Our discussion is confined to the flat band subspace via band projection, specifically the degenerate first valence band subspace. Here, we emphasize that this two-band system gives a minimal example to show the interaction-induced topologically non-trivial phases. In the strong coupling limit of the flat band systems, we can directly infer the properties of the system by diagnosing the interaction kernel because Coulomb interactions dominate and the kinetic part can be ignored. The interaction kernel analysis provides us the potential orders which can spontaneously form to lower down the total energy in the system. Taking the C_{4z} symmetric case as an example, the eigenvalues of different modes and their k space distributions are shown in Fig. B.2. Three discrete modes exhibit lower energy and are well separated from other continuous ones. The first mode, highlighted by blue dot, corresponds to the spin-polarized (SP) mode (out-of-plane ferromagnetic state) $s_z(\mathbf{k})$, while the two degenerate modes, labeled in orange, correspond to the spin-coherent modes (in-plane ferromagnetic state) $s_{x/y}(\mathbf{k})$. This indicates that Coulomb interactions favor these order parameters. Notably, the SP mode can also induce a quantum anomalous Hall effect. The C_{6z} symmetric case can also be analyzed in the same manner, see Fig. B.3.

B.4 Spin resolved Hartree Fock band structures

In this section, we present the spin resolved Hartree Fock band structures in Fig. B.4 by projecting the Hamiltonian onto six bands, including four valence bands and two

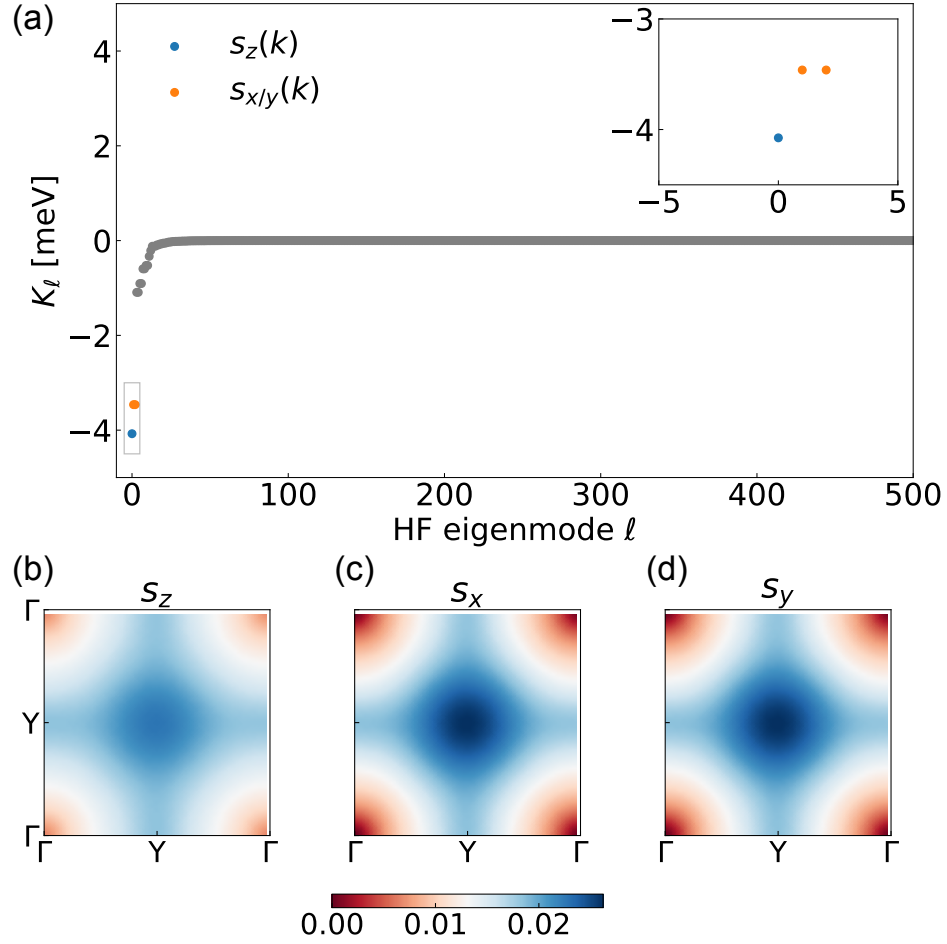


Figure B.2: (a) Eigenvalues of different modes of the Hartree Fock interaction kernel. Three discrete modes are separated from others, correspond to one spin polarized mode ($s_z(\mathbf{k})$, labeled in blue) and two spin coherent modes ($s_{x/y}(\mathbf{k})$, labeled in orange). The single particle Hamiltonian is solved on a 40×40 k -mesh when a C_{4z} symmetric potential is applied ($\delta = -5$ meV, $V_0 = 4$ meV) (b-c) The k space distribution of the Hartree Fock eigenmodes for the spin-polarized mode and two degenerate spin-coherent modes.

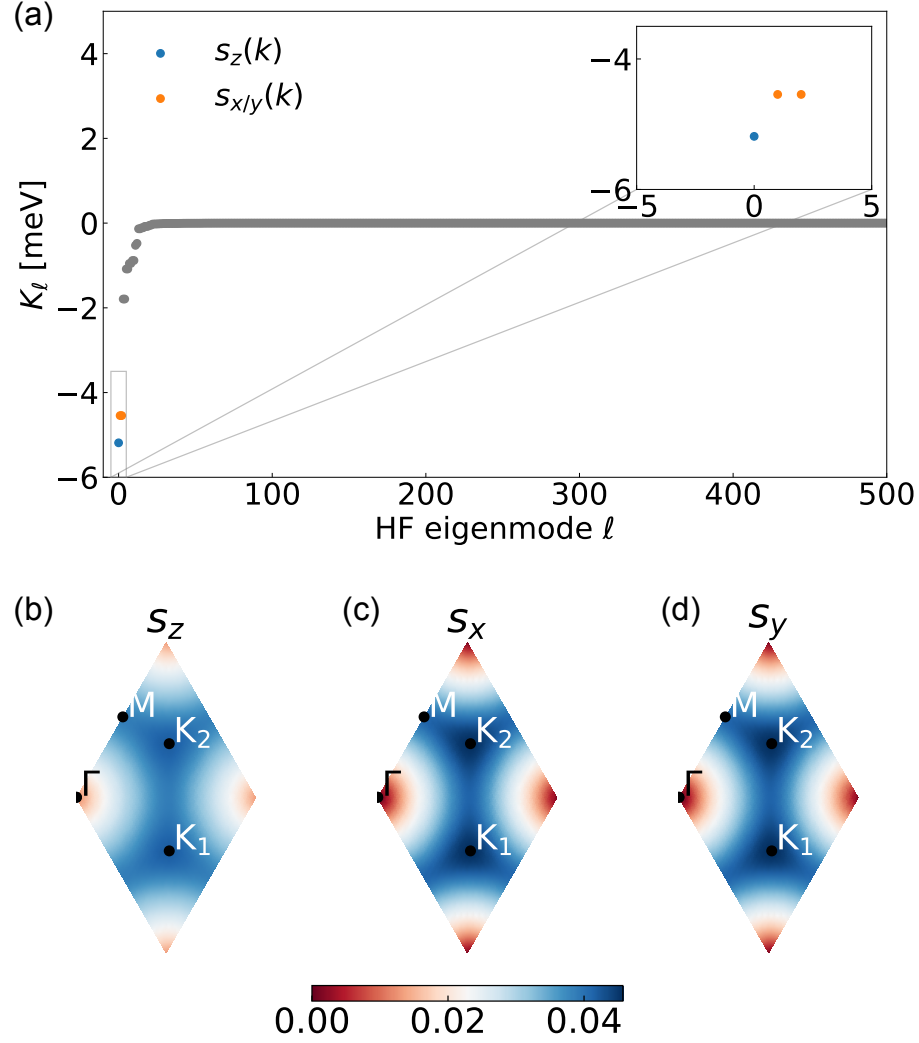


Figure B.3: (a) Eigenvalues of different modes of the Hartree Fock interaction kernel. Three discrete modes are separated from others, correspond to one spin polarized mode ($s_z \mathbf{k}$, labeled in blue) and two spin coherent modes ($s_{x/y}(\mathbf{k})$, labeled in orange). The single particle Hamiltonian is solved on a 30×30 k -mesh when a C_{6z} symmetric potential is applied ($\delta = -5$ meV, $V_0 = 3.5$ meV) (b-c) The k space distribution of the Hartree Fock eigenmodes for the spin-polarized mode and two degenerate spin-coherent modes.

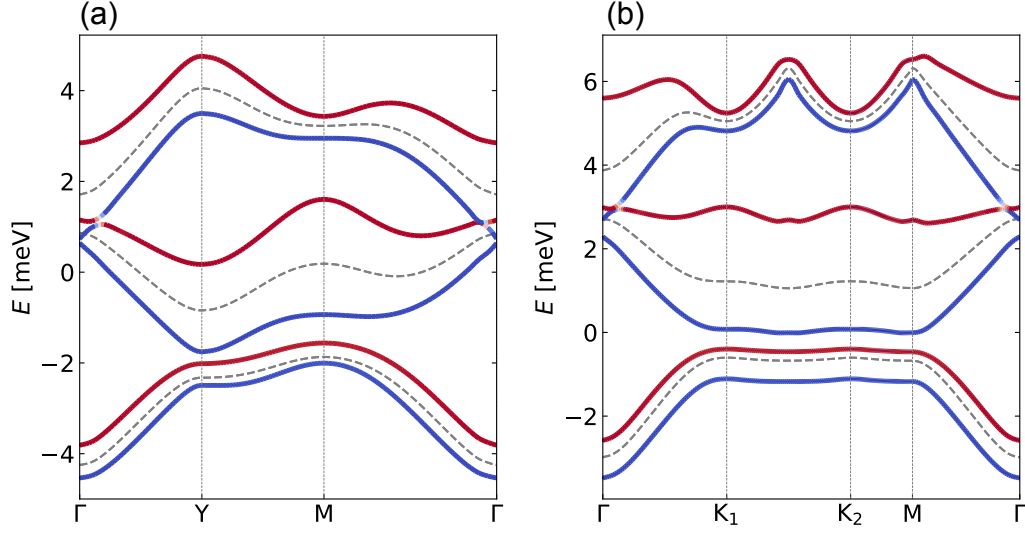


Figure B.4: (a) Six-band Hartree Fock results performed on a 20×20 k -mesh with a C_{4z} symmetric potential. ($\delta = -5$ meV, $V_0 = 4$ meV, $\epsilon = 25$) This corresponds to a anomalous Hall metal phase (AHM) due to the lack of global gap. (b) Six-band Hartree Fock results performed on a 18×18 k -mesh with a C_{6z} symmetric potential ($\delta = -5$ meV, $V_0 = 3.5$ meV, $\epsilon = 25$). This corresponds to a quantum anomalous Hall phase (QAH).

conduction bands. Panel (a) depicts the anomalous Hall metal phase (AHM) and panel (b) depicts the quantum anomalous Hall (QAH) insulator phase. The blue color labels the ‘spin up’ states while the red color labels the ‘spin down’ states. Both occupied states show the spin-polarized features.

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