Emergent fractality in graphene-based twisted van der Waals heterostructures

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EMERGENT FRACTALITY IN GRAPHENE-BASED TWISTED VAN DER WAALS HETEROSTRUCTURES

by

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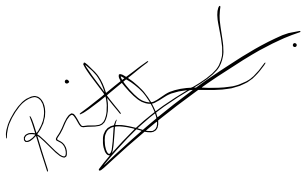
Dedicated to my family …

CERTIFICATE

This is to certify that the thesis entitled **"Emergent fractality in graphene-based twisted van der Waals heterostructures**, being submitted by **Mr. Deepanshu Aggarwal** to the Department of Physics, Indian Institute of Technology Delhi, for the award of the degree of **Doctor of Philosophy**, is a record of bonafide work carried out by him under our supervision and guidance. The results reported in this thesis have not been submitted, either in part or in full, to any other University or Institute for the award of any degree or diploma.



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ABSTRACT

Van der Waals heterostructures attracted a huge interest towards the exploration of various theoretical and experimental possibilities due to their promising tunable electronic properties. Notably, many strongly correlated phases ranging from Mott insulator to high-T_c superconductivity have been observed in two-dimensional graphene-based materials such as twisted bilayer graphene (tBLG). These correlation-driven phases result due to the non-triviality of the band structure with a strong dependence on the moiré superlattice potential. This thesis explores the complex realm of moiré patterns, a phenomenon resulting from the alignment of two layers due to twist angles and lattice inconsistencies. Exposing the tBLG to a sequence of commensurate external periodic potentials exhibits an emergent fractality, and the self-similarity of the hierarchical structures of Brillouin zones leads to a nested subband configuration inside the bandwidth of the original moiré bands, subsequently enabling a reliable low-energy band count. Furthermore, in the case of multilayer supermoiré (SM) structures created by the superposition of two or more moiré patterns from different interfaces, we demonstrate that these structures exhibit emergent fractality under specific conditions within a comprehensive low-energy framework. Mainly, through the study of hexagonal trilayer and quadlayer SM and considering that the two moiré from distinct interfaces are commensurate with each other, we derived a dominant SM length scale that is determined by the twist angles and lattice mismatches between the layers. Furthermore, we show the single-particle electronic properties along an infinitely countable set of points within a parameter space $\mathbb{R}^2$ formed by the twist-angles in two interfaces (θ_1, θ_2). To validate these theoretical predictions, we theoretically analyze modern spectroscopic techniques such as nano angle-resolved photoemission spectroscopy (nano-ARPES) intensity and the tunneling current in scanning tunneling microscopy to verify the band count and emergent fractality by using the single-particle states of the effective Hamiltonian of SM structures to obtain the momentum-space distribution of APRES intensity and local density of states for tunnelling current.

सार

वैन डेर वाल्स हेटेरोस्ट्रक्चर्स ने अपने आशाजनक ट्यूनेबल इलेक्ट्रॉनिक गुणों के कारण विभिन्न सैद्धांतिक और प्रयोगात्मक संभावनाओं के अन्वेषण की ओर भारी रुचि आकर्षित की। विशेष रूप से, ट्विस्टेड बाईलेयर ग्राफीन (टीबीएलजी) जैसी द्वि-आयामी ग्राफीन-आधारित सामग्रियों में मोट इंसुलेटर से लेकर उच्च-T_c सुपरकंडक्टिविटी तक कई दृढ़ता से सहसंबद्ध चरणों को देखा गया है। इन सहसंबंध-संचालित चरणों का परिणाम बैंड संरचना की गैर-तुच्छता के कारण होता है, जिसमें मोयर सुपरलैटिस क्षमता पर मजबूत निर्भरता होती है। यह थीसिस मोयर पैटर्न के जटिल क्षेत्र की पड़ताल करता है, जो मोड़ कोणों और जाली की विसंगतियों के कारण दो परतों के संरेखण के परिणामस्वरूप एक घटना है। टी. बी. एल. जी. को अनुरूप बाहरी आवधिक क्षमताओं के अनुक्रम में उजागर करना एक उभरती हुई अस्थिभंगता को प्रदर्शित करता है, और ब्रिलौइन क्षेत्रों की पदानुक्रमित संरचनाओं की आत्म-समानता मूल मोयर बैंड की बैंडविड्थ के अंदर एक नेस्टेड सबबैंड कॉन्फ़िगरेशन की ओर ले जाती है, जो बाद में एक विश्वसनीय कम-ऊर्जा बैंड गिनती को सक्षम करती है। इसके अलावा, विभिन्न इंटरफेस से दो या दो से अधिक मोयर पैटर्न के सुपरपोजिशन द्वारा बनाई गई बहुस्तरीय सुपरमॉयर (एसएम) संरचनाओं के मामले में, हम प्रदर्शित करते हैं कि ये संरचनाएं एक व्यापक कम-ऊर्जा ढांचे के भीतर विशिष्ट परिस्थितियों में उभरती फ्रैक्टलिटी प्रदर्शित करती हैं। मुख्य रूप से, षट्कोणीय त्रि-परत और चतुर्भुज-परत एसएम के अध्ययन के माध्यम से और यह देखते हुए कि अलग-अलग इंटरफेस से दो मोयर एक-दूसरे के अनुरूप हैं, हमने एक प्रमुख एसएम लंबाई स्केल प्राप्त किया जो परतों के बीच मोड़ कोण और जाली बेमेल द्वारा निर्धारित किया जाता है। इसके अलावा, हम दो इंटरफेस (θ_1, θ_2) में ट्विस्ट-एंगल द्वारा बनाए गए पैरामीटर स्पेस $\mathbb{R}^2$ के भीतर अनंत रूप से गणना योग्य बिंदुओं के सेट के साथ एकल-कण इलेक्ट्रॉनिक गुणों को दिखाते हैं। इन सैद्धांतिक भविष्यवाणियों को मान्य करने के लिए, हम सैद्धांतिक रूप से आधुनिक स्पेक्ट्रोस्कोपिक तकनीक जैसे नैनो-एंगल-रिज़ॉल्व्ड फोटोमिशन स्पेक्ट्रोस्कोपी (एआरपीईएस) की तीव्रता और स्कैनिंग टनलिंग माइक्रोस्कोपी में टनलिंग करंट का विश्लेषण करते हैं ताकि बैंड की गिनती और एसएम संरचनाओं के प्रभावी हैमिल्टनियन की एकल-कण अवस्थाओं का उपयोग करके उभरते फ्रैक्टलिटी को सत्यापित किया जा सके।

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LIST OF SYMBOLS

$\mathbb{N}$ The natural numbers

$\mathbb{Z}$ The integer numbers

$\mathbb{Q}$ The rational numbers

$\mathbb{R}$ The real numbers

$\mathbb{C}$ The complex numbers

$\in$ Set memberhood

$\mathbb{R}^n$ Space of n-tuples of real numbers

$\|x\|$ Norm of a vector x

LIST OF ACRONYMS/ABBREVIATIONS

1D one-dimensional

2D two-dimensional

3D three-dimensional

SLG single layer graphene

hBN hexagonal Boron Nitride

BLG bilayer graphene

BZ Brillouin zone

FBZ first Brillouin zone

DOS density of states

VHS Van Hove singularity

SL superlattice

mBZ moiré Brillouin zone

tBLG twisted bilayer graphene

MATBLG magic-angle twisted bilayer graphene

vdW van der Waals

MF moiré fractal

tDBG twisted double bilayer graphene

tTLG twisted trilayer graphene

TMD transition metal dichalcogenide

G-G Graphene/Graphene

mEP moiré external potential

G-hBN Graphene/hexagonal Boron Nitride

D_f fractal dimension

hBN-G-hBN hBN encapsulated single layer graphene

hBN-BLG-hBN hBN encapsulated bilayer graphene

SM supermoiré

SMS supermoiré structure