

ABSTRACT

The discovery of graphene in 2004 has started extensive research in two-dimensional (2D) materials due to their potential applications in next-generation electronics and quantum devices. The 2D van der Waals (vdW) heterostructures formed by stacking atomically thin layers provide a powerful platform for engineering novel quantum phenomena. Twisted bilayer graphene (TBLG) forms when two graphene layers are rotated by twist angle (θ) relative to each other, forming a moiré superlattice. The resulting electronic and vibrational properties depend sensitively on the twist angle and interlayer coupling. While extensive studies have focused on the magic-angle ($\theta \approx 1.1^\circ$) TBLG, where flat electronic bands lead to correlated insulating and superconducting states, large-angle TBLG ($\theta > 3^\circ$) remains comparatively less explored. For this regime of large twist angles, low energy electronic bands retain linear dispersion with a Fermi velocity close to that of monolayer graphene, yet interlayer coupling and electron-electron interactions can still give rise to rich interaction-driven phenomena. This thesis focuses on investigating the interplay between lattice vibrations, electronic structure, and quantum Hall transport in large-angle TBLG through Raman spectroscopy and low-temperature magnetotransport measurements.

The first part of the thesis investigates twist-angle-dependent interlayer coupling in TBLG using Raman spectroscopy. Systematic Raman studies reveal a resonance enhancement of the G band arising from interband parallel transitions that occurs when the laser excitation energy matches the energy separation between conduction and valence band van Hove singularities. The evolution of the 2D peak indicates progressive electronic decoupling of the graphene layers with increasing twist angle. The appearance of the R and R' modes provide direct signatures of moiré potential-induced intra- and intervalley scattering processes. Furthermore, the behavior of Raman combination modes is used to identify a commensurate-incommensurate transition in TBLG, which originates from the reduction of interlayer coupling and increased compressive strain at larger twist angles. The second part of this thesis presents a detailed study of magnetotransport in large-angle TBLG in the quantum Hall regime. Layer-resolved Shubnikov-de Haas oscillations are analyzed using a two-layer electrostatic model to extract the carrier densities in the individual graphene layers. These measurements reveal that charge screening and electrostatic imbalance between the layers significantly modify the Landau level structure, leading to systematic Landau level crossings. Transport measurements further show the interaction-driven lifting of spin, valley, and layer degeneracies, resulting in the emergence of broken-symmetry integer quantum Hall states in TBLG. Temperature-dependent measurements show that these states weaken and disappear with increasing temperature, confirming their many-body origin. The final part of this thesis investigates quantum Hall ferromagnetism and interaction-driven phase transitions in large-angle TBLG. Transport measurements near the charge neutrality point reveal a Kosterlitz-Thouless-type transition into a quantum Hall insulating state associated with intervalley-coherent (Kekulé) order under high magnetic fields. Tilted-field measurements further indicate that the low-energy excitations of the broken-symmetry states are governed by collective spin and valley skyrmion textures. Pronounced hysteresis observed in longitudinal and Hall resistances establishes the presence of first-order phase transitions between competing quantum Hall ferromagnetic states. Using a dual-gated device, we demonstrate that the displacement field acts as an effective tuning parameter to control the layer polarization and stabilize ferromagnetic phases, leading to sweep-direction-dependent hysteresis associated with skyrmion-mediated domain walls. Altogether, this thesis highlights the role of moiré-engineered interlayer coupling and electron-electron interactions in determining the emergent electronic behaviour of TBLG and establishes large-angle TBLG as a tunable platform to explore correlated quantum phenomena in 2D materials.

Keywords: twisted bilayer graphene, Raman spectroscopy, broken-symmetry states, Quantum Hall Ferromagnetism, skyrmions