様式３

論　文　内　容　の　要　旨

氏　名　（　城 谷 宇 映　）	
論文題名	Theory of nonlinear optical responses in moiré materials モアレ物質における非線形光学応答の理論

In the present thesis, we have performed a systematic theoretical study of nonlinear optical responses in moiré materials. Moiré materials, formed by stacking two-dimensional crystals with a relative twist or lattice mismatch, generate long-wavelength superlattices that strongly reconstruct electronic band structures. This reconstruction can produce moiré flat bands, providing a highly tuneable platform for exploring quantum phenomena. In parallel, nonlinear optical responses have emerged as powerful probes of electronic properties, owing to their sensitivity to symmetry, band topology, and quantum geometry beyond linear-response measurements.

In this work, we investigate the second-order optical responses in twisted double bilayer graphene (TDBG), a moiré system composed of two AB-stacked bilayer graphene sheets twisted relative to each other. Depending on the relative orientation of the bilayers, TDBG realises two stacking variants, AB-AB and AB-BA, which possess different symmetries while hosting very similar electronic band structures. We focus in particular on the shift current and second-harmonic generation (SHG), which are frequency-mixing responses allowed only in noncentrosymmetric crystals. The shift current is a rectified dc photocurrent generated under illumination and is closely connected to band topology through the Berry connection, while SHG is the complementary frequency-doubling process shaped by the coexistence of one-photon and two-photon excitation channels. Moiré flat bands enhance the joint density of states and can significantly amplify these nonlinear optical signals.

Using TDBG as a concrete example, we perform a systematic theoretical analysis of the shift current and SHG as functions of the twist angle, Fermi level, vertical bias voltage, and stacking configuration. We find that both responses are strongly enhanced as the twist angle is reduced. This enhancement originates from the formation of moiré flat bands, which concentrate low-energy interband transitions within a narrow energy window, leading to a large joint density of states. In the case of SHG, we further identify an enhancement mechanism arising from an abundance of a double-resonant processes enabled by the dense spectrum of moiré flat bands. Unlike in non-moiré systems, TDBG supports simultaneous one-photon and two-photon excitation channels over wide regions of the moiré Brillouin zone, resulting in a qualitatively distinct resonant enhancement of SHG.

We also find that second-order optical responses in TDBG exhibit a pronounced sensitivity to the Fermi energy. In the low-frequency regime, the response is dominated by transitions between neighbouring moiré flat bands. As the Fermi level is tuned across successive band edges, the dominant contributing transitions change abruptly, giving rise to sign reversals in the shift current and corresponding phase shifts in SHG. This behaviour contrasts with the smooth carrier-density dependence typical of conventional materials and reflects the reconstructed band structure characteristic of moiré systems.

Finally, we uncover a systematic relation between the nonlinear optical responses of the two stacking variants of TDBG, AB-AB and AB-BA. At large vertical bias, the shift current below charge neutrality exhibits opposite signs in the two configurations, corresponding to a relative π-phase shift in SHG. By analysing an uncoupled TDBG model in which moiré coupling is switched off, we show that this behaviour can be qualitatively understood as a consequence of the 180-degree relative rotation of one bilayer, which reverses the in-plane polarisation. This stacking-dependent signature remains visible even after incorporating the full moiré-induced band reconstruction, highlighting the importance of stacking geometry in determining second-order optical responses in TDBG.

Taken together, these findings clarify the role of moiré band reconstruction and stacking geometry in second-order optical signals, and offer a systematic perspective on engineering nonlinear optical responses in moiré materials.