Hybridization of Moiré States in Twisted Bilayer hBN

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Twisted van der Waals structures attracted immense attention in recent years. Often, these systems invoke the concept of strongly localized excitations (quasiparticles (QP's)), i.e. charged electrons, holes, or excitons, subject to a periodic potential landscape whose energy and spatial distribution is determined by the twisting angle applied. Guiding the development of moiré platforms necessitates quantitative theoretical understanding of their properties, however, computing these systems from first principles is a challenging task since large supercells with thousands of atoms are necessary to properly describe the moiré superlattice. Typically, a local approximation is taken where the QP's in the twisted bilayers are subject to a local potential landscape formed by just the distinct stacking orders in the 2D lattice. This picture is invoked e.g., in the description of effective exciton potentials from the variation of the local band gaps. To what extent the non-local long-range interactions modify this picture and how the individual QPs (de)localize in the moiré potential, however, remains unknown. Moreover, calculations are limited to mean-field methods and the role of non-local correlations on the spatial extent of Dyson orbitals are, therefore, also unknown.

Here, we overcome these computational difficulties and investigate the QP landscapes by employing the GW approximation in the stochastic many-body perturbation theory (MBPT) formulation. We extend the technique for determining the Dyson orbitals via a stochastic compression, enabling calculations of QP states in large nanoscale systems. For simplicity, we investigate twisted hexagonal boron nitride (t-hBN) and consider four small twist angles: 2.45°, 2.86°, 3.48° and 5.09°, with structures containing up to ~18,000 electrons. Our results reveal how localized moiré states hybridize with each other and lead to a nontrivial interplay between local structure and non-local correlations. We find that the local gap variation is greatly exaggerated, significantly changing the excitonic potential. For most twist angles, the non-local interactions lead to band-edge QP state mixing (hybridization), which is suppressed for angles around 3.48°. This behavior is mostly determined by the strength of interactions and the localization in the high symmetry regions of the moiré cell.