Title: Bethe Salpeter equation calculations in moiré superlattices with thousands of atoms in the unit-cell

Abstract:
Recent experimental measurements have demonstrated signatures of novel exciton states in the moiré superlattices of transition metal dichalcogenide bilayer heterostructures. However, the microscopic nature of these moiré excitons was not well understood, and previous studies relied often on empirically fit models. Brute force first-principles *GW*-Bethe Salpeter equation (GW-BSE) calculations are computationally intractable due to thousands of atoms in the reconstructed moiré unit-cell. We present a novel computational approach, the pristine unit-cell matrix projection (PUMP) method [1] which makes it possible to accurately study excitons in large-area transition metal dichalcogenide moiré superlattices. Through these calculations, we discovered a rich diversity of excitonic states. In rotationally aligned WSe_2/WS_2 moiré superlattice, we find some excitons of a modulated Wannier character and others of a previously unidentified intralayer charge-transfer character [1]. In 57.7° twisted bilayer WS_2, we discover layer-hybridized excitons with in-plane charge transfer character. These characteristics originate from the strong modulation of electron wavefunctions due to atomic reconstructions of the superlattice. Experimental reflection contrast [1], electron energy loss spectroscopy [2] and scanning tunneling spectroscopy measurements confirm these predictions. The PUMP approach can also be extended to study optical spectrum of shallow defects in materials, which also require large supercell calculations.

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