Interlayer Exciton Diffusion in Twist-Angle-Dependent Moiré Potentials of WS_2-WSe_2 Heterobilayers

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The nanoscale periodic potentials introduced by so-called moiré patterns in semiconducting van der Waals (vdW) heterostructures provide a new platform for designing exciton superlattices. To realize these applications, a thorough understanding of the motion of the excitons in the moiré potentials is necessary. Here, we investigated interlayer exciton dynamics and transport modulated by the moiré potentials in WS_2/WSe_2 heterobilayers in time, space, and momentum domains using transient absorption microscopy combined with first-principles calculations. Exciton transport deviates from normal diffusion, due to the interplay between the moiré potentials and strong many-body interactions, leading to exciton-density- and twist-angle-dependent behavior. Experimental results verified the theoretical prediction of energetically favorable K-Q interlayer excitons and unraveled exciton-population dynamics that was controlled by the twist-angle-dependent energy difference between the K-Q and K-K excitons. These results have important implications for designing vdW heterostructures for exciton and spin transport as well as for quantum communication applications.
The studied heterobilayers have two stacking orientations with twist angles of θ = 0° and 60°, which are energetically favorable in the modified two-step CVD growth. Both structures have type-II band alignment, resulting in the formation of spatially-indirect interlayer charge-transfer excitons, with electrons and holes residing in the WS_2 and WSe_2 layers, respectively. The lowest-energy transition is always K-Q and therefore K-Q interlayer excitons are expected to represent the ground state instead of the more commonly discussed K-K excitons. The spatial variations of the moiré potential for 0° are much stronger (deep potential) than for 60° (shallow potential). Thus, two predictions could be made based on the DFT calculations: (i) the population dynamics of the K-K and K-Q excitons should be affected by the twist-angle-dependent energy difference between the two transitions; and (ii) the twist-angle-dependent moiré potentials should lead to different degrees of localization of the interlayer excitons in both systems. The validation of both predictions will be discussed in this presentation.
The localization and delocalization of the interlayer excitons presented here have important implications for the potential applications of heterostructures: for long-range transport, more delocalized interlayer excitons are preferred and, therefore, deep moiré potentials should be avoided. On the other hand, for applications, such as, quantum emitters, deep moiré potential should be preferred, to localize excitons. We also stress that K-Q interlayer excitons are the ground state instead of the commonly assumed K-K excitons and should be considered when discussing interlayer excitons in the WS_2/WSe_2 systems.