Twist Angle Dependent Phonon Hybridization in WSe_2/WSe_2 Homobilayer

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The emerging moiré superstructure of twisted Transition Metal Dichalcogenides (TMDs) leads to various correlated electronic and optical properties compared to twisted bilayer graphene. In such versatile architecture, phonons can also be renormalized and evolve due to atomic reconstruction, which in turn, depends on twist angle. However, observing this reconstruction and its relationship to phonon behaviour with conventional, cost-effective imaging methods remains challenging. Here, we used non-invasive Raman spectroscopy on twisted WSe_2/WSe_2 homobilayers to examine the evolution of phonon modes due to interlayer coupling and atomic reconstruction. Unlike in the natural bilayer (NB), 0^0 as well as 60^0 devices; the nearly degenerate A_{1g}/E_{2g} mode in the twisted samples (1^0-7^0) split into a doublet of A_{1g}/E_{2g}^+ and E_{2g}^- in addition to the non-degenerate B_{2g} mode, and maximum splitting is observed around 2^0-3^0. Our detailed theoretical calculations qualitatively capture the splitting and its dependence as a function of the twist angle and highlights the role of the moiré potential in phonon hybridization. Additionally, we found that for 2^0 twisted device, the anharmonic phonon-phonon interaction is higher than the natural bilayer and decreases for larger twist angles. Interestingly, we observed anomalous Raman frequency softening and line-width increase with decreasing temperature below 50 K, pointing to the combined effect of enhanced electron-phonon coupling and cubic anharmonic interactions in moiré super-lattice.

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