Renormalization of phonons and excitons in twisted heterostructures of transition metal dichalcogenides

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Heterostructures of two-dimensional transition metal dichalcogenides have been extensively studied as the alignment of the bands in the constituent materials allow for manipulation of optoelectronic and transport properties. Several factors such as the band offset between the bands of the two constituent materials and the twist angle between the crystallographic directions of the two layers determine the physical properties of these structures. It has been demonstrated that the phonon spectrum as well as the electronic band structure and optoelectronic properties change systematically as the twist angle between two layers of a given set of materials is varied. Furthermore, at very small twist angles, atomic-scale lattice reconstruction [1] is observed and should be accounted for in describing the physical properties of heterostructures. Recently, we found that moiré superlattices made from single layers of MoS_2 and WSe_2 with a fairly large lattice mismatch (~4.5%) exhibit a pair of torsional distortions with opposite chirality irrespective of the twist angle [2]. The whirlpool-shaped periodic lattice distortions introduce fuzziness in the Raman spectra and universal redshifts to the intralayer excitons for all twist angles. Both of these modulations become weaker as the twist angle increases but do not disappear, whereas they are turned off when the constituent layers are not tightly coupled. In a lattice matched (~0.24%) heterostructures of $MoSe_2$ and WSe_2, a series of moiré phonons are observed, and the low-frequency Raman spectra show a rich array of interlayer shear and breathing modes that evolve with the twist angle [3]. Furthermore, the interlayer excitons that appear strong in the twisted heterostructures with the twist angles near 0° or 60° have different energies and photoluminescence excitation spectra for the two cases, which results from different electronic structures and carrier relaxation dynamics. These results demonstrate that the details of the lattice interactions as well as the twist angle should be considered in designing heterostructure-based devices.

References

[1] H. Yoo, *et al*., Nature Materials, 18 (2019) 448-453.
[2] J. Kim, et al., Nature Materials, 21 (2022) 890-895.
[3] S.Y. Lim, et al., ACS Nano, 17 (2023) 13938-13947.