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Raman spectroscopy of twisted bilayer MoS₂

Compared to conventional semiconductor heterostructures, van der Waals heterostructures (vdWHs) based on two-dimensional materials (2DMs) are readily fabricated by direct chemical-vapor-deposition growth or wet/dry transfer. Those vdWHs have ultraclean and atomically sharp interfaces, providing a versatile platform for studying interface physics. Also, the material choice of the components, layer thickness and interlayer twist angle θ widely enrich the vdWHs and provide additional degrees of freedom to engineer their optical and electronic properties. The Moiré patterns in vdWHs create a periodic potential for electrons, excitons and phonons to yield many interesting phenomena, such as Hofstadter butterfly spectrum and Moiré excitons. Here, in the twisted bilayer MoS₂ (tBLM), one of the simplest prototype of vdWHs, we show how the periodic potentials of Moiré patterns induce different phonon modes and relate those to the lattice dynamics from its constituent, monolayer MoS₂. We report the observation of new Raman modes related to Moiré phonons in asgrown/transferred tBLMs with different twist angles, which are folded from the off-center phonons in monolayer MoS₂. However, the folded phonons related to crystallographic superlattices are not observed in the Raman spectra,[1] By varying the twist angle, the Moiré phonons of tBLM can be used to map the phonon dispersion of the constituent layers (Fig.1). The lattice dynamics of the Moiré phonons are modulated by the patterned interlayer coupling resulting from periodic potential of Moiré patterns, as confirmed by density functional theory calculations. The Raman intensity related to Moiré phonons in all tBLMs are strongly enhanced when the excitation energy approaches the C exciton energy. This study can be extended to investigate Raman spectra in various vdWHs to deeply understand their Moiré phonons, lattice dynamics, excitonic effects and interlayer coupling.

References

[1] Lin, Miao-Ling, Tan, Qing-Hai, Wu, Jiang-Bin, Chen, Xiao-Shuang, Wang, Jin-Huan, Pan, Yu-Hao, Zhang, Xin, Cong, Xin, Zhang, Jun, Ji, Wei, Hu, Ping-An, Liu, Kai-Hui, Tan, Ping-Heng, ACS Nano, 12(2018), 8770

Figure

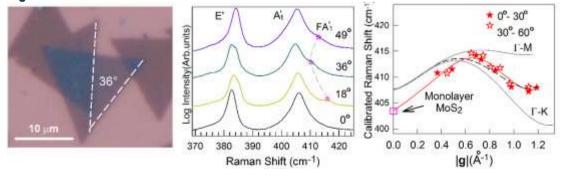


Figure 1 The optical images and Raman spectra of tBLM in the A'₁ spectral region by varying the twist angle. The wavevector-dependent frequencies of Moiré phonon related to mode are summarized, along with the theoretical phonon dispersion of A'₁ mode along the Γ -K and Γ -M directions are shown.