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Exciton Band Structures of TMDs measured by Momentum resolved EELS

Abstract

Two-dimensional semiconducting transition metal dichalcogenides (TMDs) are continually attracting extensive research interest in their excitonic, valleytronic, photonic and catalytic applications owing to their unique electronic properties and defect physics. Besides the well-known indirect-to-direct transition crossover when the thickness decreases to monolayer limit, the weakly-allowed spin-flipping involved electronic transition appears as dark exciton in the out-of-plane polarized optical pumping[1] (grazing incidence) or under the exposure of near-field surface plasmon polariton[2,3]. Benefitting from the inevitable vast defects or introduced defect engineering of TMDs, these defective sites act as active reaction centers in catalysis or color centers in the quantum single photon emission[4,5] in semiconducting TMDs.

Here we employ momentum-resolved electron energy loss spectroscopy (EELS) in a transmission electron microscope (TEM) to probe the momentum dependent low loss fine structures (excitons, interband transitions and plasmons) of TMDs. With the help of a double Wien filter monochromator, the energy resolution can be improved down to ~30 meV, which enables the exploration of exciton and even phonon band structure. High momentum resolution is achieved in the diffraction mode using parallel beam illumination in a TEM. Different dispersion structures were observed for the various excitons in monolayer TMDs. Subtle in-plane anisotropy in the valence electron excitation is found, especially in the band-edge transition excitons. Complicated exciton structures are unambiguously identified and assigned with their specific origins through the momentum dependence of their oscillator strength. Through tracking the bandgap evolution by EELS, we'll also show the asymmetric response of valance band extrema and conduction band extrema to the variation of thickness of TMDs.

References

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Figures

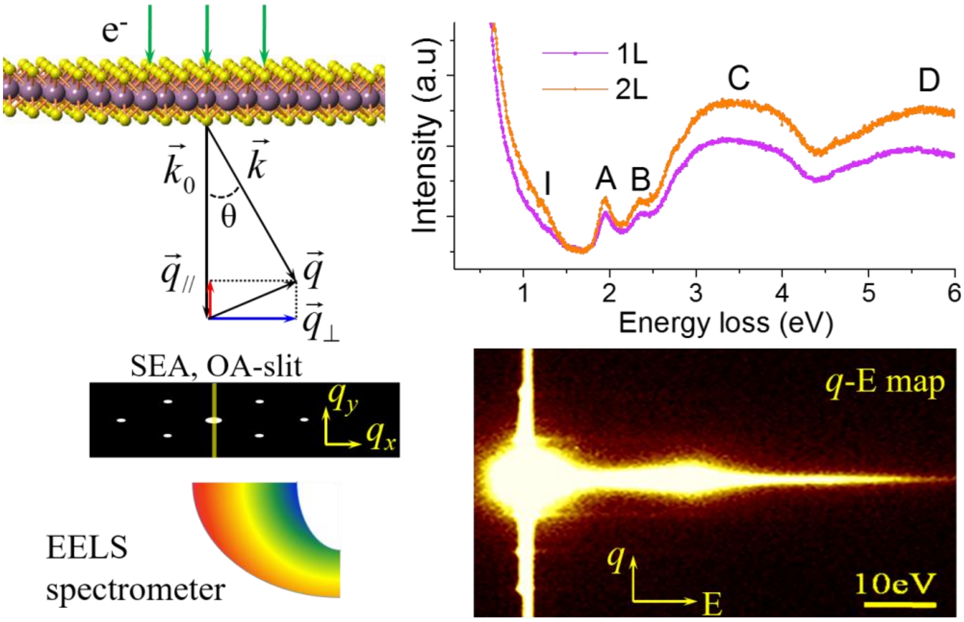

Figure 1: Momentum resolved EELS to reveal the band structure of excitons in TMDs. Momentum *q* is selected in the diffraction space to obtain *q* dependent low loss fine structures.