

Exploring the Bright Side and the Dark Side of Excitons in Atomically-thin Transition Metal Dichalcogenides

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In this talk, we will explore fundamental properties of bright and dark exciton transitions in Transition Metal Dichalcogenides (TMDs). On the bright side, we will probe a simple question: can TMDs act as an atomically-thin mirror at the bright exciton resonance? Such an experimental realization places a stringent demand on the excitons – the radiative linewidth should dominate the total transition linewidth, a criteria rarely fulfilled in solid-state systems. Remarkably, we observe resonant reflection of up to 85% of incident light from a monolayer of MoSe₂, indicating that the radiative decay rate of excitons can be up to an order of magnitude larger than the non-radiative and dephasing rates. Furthermore, we demonstrate that these mirrors can be electronically switched, and exhibit strong power- and wavelength-dependent hysteresis. On the dark side, because of strong spin-orbit coupling the lowest-energy excitonic states in some TMD monolayers (such as WSe₂ and WS₂) involve nominally spin-forbidden optical transitions and are thus optically dark. Probing and understanding these dark excitons provides crucial insights into the fundamental properties of TMD monolayers, however their presence typically can only be inferred through indirect measurements. We will introduce a method for probing the optical properties of two-dimensional (2D) materials via near-field coupling to surface plasmon polaritons (SPPs). This coupling selectively enhances optical transitions with dipole moments normal to the 2D plane, enabling direct detection of dark excitons in TMD monolayers. When a WSe₂ monolayer is placed on top of a single-crystal silver film, its emission into near-field-coupled SPPs displays new spectral features whose energies and dipole orientations are consistent with dark neutral and charged excitons.