Ultrafast laser control of magnetization switching in two-dimensional van der Waals heterostructures

Photo-induced excited state dynamics in two-dimensional van der Waals heterostructures

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In Short

- Ground state properties of 2D magnets and vdW heterostructures
- Screening and understanding the fundamental electronic and excitonic properties of 2D vdW heterostructures
- Explore the effects of external stimuli and many-body interaction on excited state dynamics behavior of 2D vdW heterostructures

The major goal of this project is to explore control of light-matter interactions in 2D van der Waals (vdW) heterostructures constituted by well-understood vdW single layers. Stacking various 2D single layers to vdW heterostructures not only retains the outstanding properties of single components but also introduces numerous novel physics characteristics that do not exist in 2D single layers. Recently, the state-of-the-art development of photo-induced excited state dynamics in such 2D systems has become a salient aspect of modern energy conversion and optoelectronic devices [1-3].[Fig. 1] In experiments, the photogenerated carrier relaxation lifetime in typical transition-metal dichalcogenides can reach hundreds of picoseconds. Such photo-induced dynamics behavior is believed to be significantly modulated by various quasiparticle interactions and external stimuli while it has scarcely been studied previously. More importantly, the clear understanding and effective control of the real-time excited state dynamics behavior in 2D systems remain largely unexplored and unknown.

Project objectives This research project aims to explore the photo-induced excited state dynamics properties of vdW heterostructures constituted by well-understood 2D vdW monolayers, where the heterostructures are yet to be tuned for diverse device applications. The successful isolation of both semiconducting (MoS_2, In_2Se_3, $FePS_3$, etc.) and metallic (graphene, VS2, etc.) 2D vdW sheets provides a fertile arena for interface and device engineering. The nature of chemistry, intercalants, orientation, and strain at their interfaces, as well as quasiparticle interaction, remain less explored from a fundamental

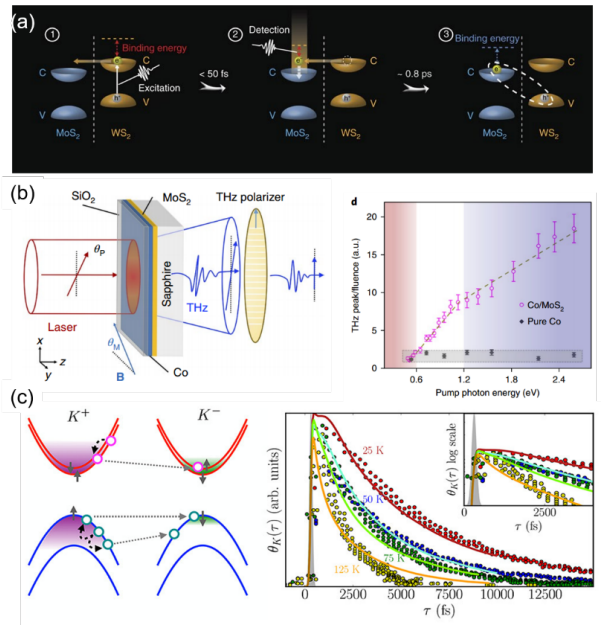

Figure 1: *(a) Charge separation processes in the MoS_2/WS_2 heterostructure. (b) Schematic of Co/MoS_2 heterostructure and experimental configuration. (THz peak)/(absorbed fluence) of our devices under different pump wavelengths. (c) Schematic representation of the selection rules for circularly polarized light at the K± valleys. Kerr angle $\theta_K(\tau)$ for several temperatures. The shadow gray region represents the pump pulse.[1-3]*

perspective, providing us a unique opportunity to understand the effect of these external stimuli and many-body interaction on photo-activated excited state dynamics in different 2D heterostructures.

Screening and understanding the fundamental electronic and excitonic properties of 2D vdW heterostructures

The first phase of the project will be focused on screening suitable heterostructures and understanding the ground state and excited state properties. Potential 2D semiconductor/semi-conductor or semiconductor/metal heterostructure systems will be evaluated from their band alignments, band gaps, band edge positions, feasibility as well as environmental and structural stability. We will consider different stacking configurations and find the energetically most stable structures.

Next, our goal will be to explore the exciton properties in these heterostructures. To consider that, DFT calculations combined with GW approximation plus Bethe-Salpeter equation (GW+BSE), have to

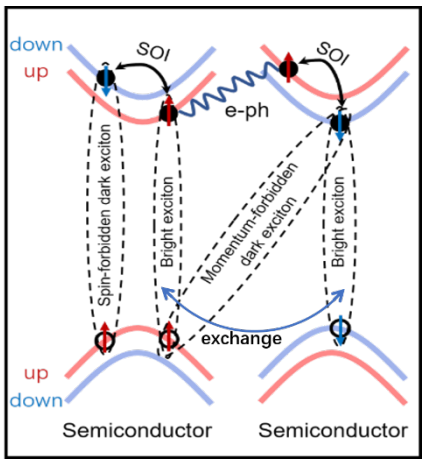

Figure 2: *Schematic diagram of the interlayer exciton transition and bright-to-dark exciton transition processes are shown. They are induced by e-h exchange, spin-orbital interaction (SOI), and e-ph coupling, respectively.*

be performed. In general, the electrons in each layer can be individually excited by an ultrafast laser pump to form independent intralayer excitons with strong exciton binding energies, exhibiting apparent exciton luminescence, i.e. bright exciton. Meanwhile, the electrons can also hop from one layer to another directly under light illumination, or through photogenerated charge transfer and hot carrier relaxation, to form interlayer excitons. This exciton belongs to dark exciton, which is not directly accessible by optical techniques. We will also consider the spin-orbit coupling (SOC) effect in the transition dipole moment calculations. And the modulation of the spin-flip between the spin-up and spin-down channel will be studied, aiming to reveal bright (carriers reside in the same spin channel) or dark (carriers reside in the different spin channel) spin exciton in the complex heterostructures.[Fig. 2]

Explore the effects of external stimuli and many-body interaction on excited state dynamics behavior of 2D vdW heterostructures

The second phase will involve systematic approaches to explore the effect of external stimuli on the 2D vdW heterostructures. In this phase, various surface or interface modifications, such as doping, intercalation, defect formation, physisorption, and chemical functionalization, will be used to systematically vary the electronic or excitonic properties. For example, by changing the doping atom or the ligand we can change the charge distribution, and modulate the band gap and overlap of frontier orbitals, thus affecting the excited state dynamics behavior. In addition, we will consider constructing various quasiparticles in target heterostructures and explore their interaction and influence on excited state dynamics processes. We also consider the application of laser pumps to determine and monitor the real-time dynamics of the various photo-induced excited state products and modulate these dynamics processes [4, 5]. We propose to optically manipulate the carrier/exciton/spin transfer and recombination in 2D vdW heterostructures using time-dependent all-electron full-potential methods, aiming to reveal how the laser pump induces charge transfer in these heterostructures.

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More Information

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