Strong-Coupling Ultrafast Charge Transfer in MoS_2/WS_2 Bilayers

Since the discovery of graphene and the rise of MoS_2, atomically thin two-dimensional (2D) crystals have grown into a huge family of materials ranging from semimetal, semiconductors to insulators. Monolayer transition-metal dichalcogenides denoted as MX_2 (*e.g.*, M=Mo, W, and X=S, Se, Te) have been prepared by physical exfoliation and chemical vapor deposition, providing more choices for 2D materials. The MX_2 materials share similar crystalline structures and symmetries, but possess distinct electronic properties in band gaps and photoabsorption. The heterostructures vertically reassembled from different two-dimensional materials form even richer material systems, and thus provide a new platform for investigating new physics and exploring new applications. The heterostructures of two types of MX_2 materials are of particular interest, because many of them form type II heterojunctions, which facilitate the efficient separation of photo-excited electrons and holes and therefore exhibit great potentials in the applications of photodetectors, photovoltaic cells and light emitters.

Light-induced interlayer ultrafast charge transfer in two-dimensional heterostructures provides a new platform for optoelectronic and photovoltaic applications. Hong *et al*. firstly reports that collective motion of excitons at the interface leads to plasma oscillations associated with optical excitation in the ultrafast charge transfer in such van der Waals heterostructures, providing a good insight in this new phenomenon (Hong *et al.*, *Nat. Nanotechnol*. 2014, 9, 682). The charge separation process is generally hypothesized to be dependent on the interlayer stackings and interactions; however, the quantitative characteristics and detailed mechanism remain challenging and elusive.

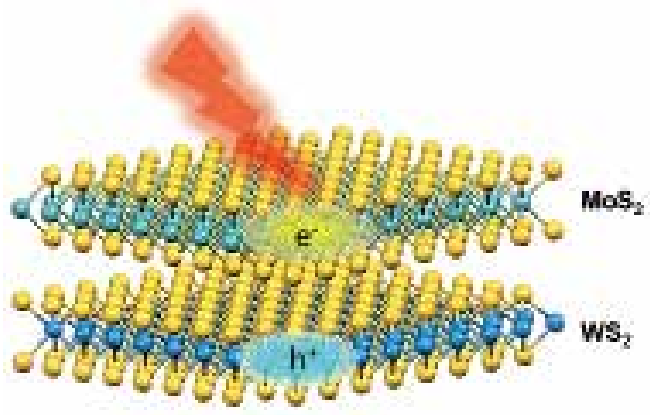
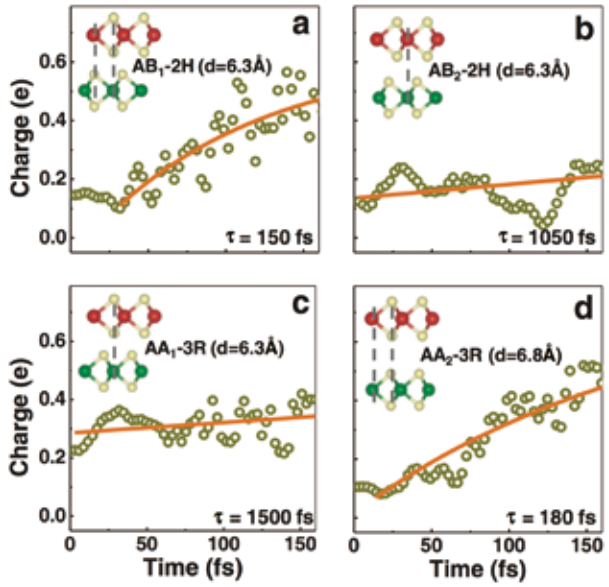


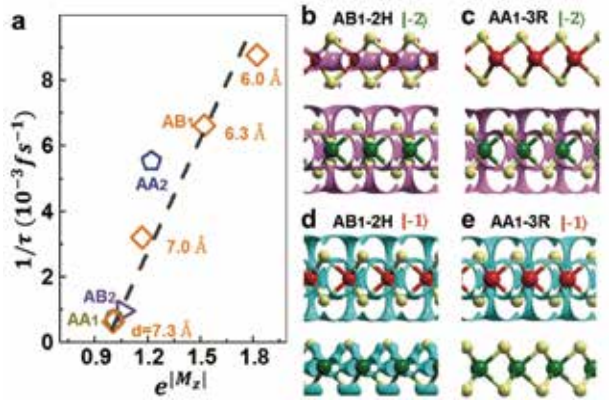
Illustration of a MoS_2/WS_2 heterostructure and ultrafast charge dynamics. (Image by Institute of Physics)



Hole transfer evolution for MoS_2/WS_2 in different stacking configurations. (Image by Institute of Physics)

Now a group from the Institute of Physics, Chinese academy of sciences led by Prof. MENG Sheng demonstrates systematically the stacking-dependent interlayer charge transfer in MoS_2/WS_2 bilayer model system by first-principles time-dependent density-functional theory simulations. They show that the slight change in interlayer geometry can significantly modulate the charge transfer time from 100 fs to >1 ps scale. Detailed analysis further reveals that the transfer rate in MoS_2/WS_2 bilayers is governed by the electronic coupling between specific interlayer states, rather than interlayer binding strength, and follows a universal dependence on the specific-state-coupling strength. Surprisingly, this dependence on the coupling strength breaks the Fermi's golden rule, implying that the bilayer system is in the strong-coupling regime beyond the scope of perturbative theory. Their results establish the interlayer stacking as an effective freedom to control ultrafast charge transfer dynamics in 2D heterostructures, facilitating their future applications in optoelectronics and light harvesting.

This study entitled "Interlayer-State-Coupling Dependent Ultrafast Charge Transfer in MoS2/WS2 Bilayers" was published in *Advanced Science*.



Interlayer-state-coupling dependent charge transfer rate in MoS_2/WS_2 bilayers.

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