Good Defects at Interfaces: Exploiting Defects at Van der Waals Interfaces between Layered Materials towards Efficient Photodetectors

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Abstract

Van der Waals (vdW) materials, such as graphite and transition metal dichalcogenides (TMDs, e.g. WS_2), are layered materials made up of strong two-dimensional (2D) sheets that are weakly bound in the third dimension through vdW forces. Owing to the weak interlayer interaction, atomically thin vdW monolayers can be produced via mechanical exfoliation. The most notable example is graphene, a monolayer of graphite, produced by peeling off layers from the bulk using the simple scotch tape method.[1] Graphene is a semimetal, and exhibits extremely high charge carrier mobility, making it relevant for electrical devices. Other vdW monolayers, e.g. those exfoliated from TMDs, are usually semiconductors which are also useful for many optical and optoelectronic applications. 2D monolayers from different materials can be stacked vertically to form so-called "vdW heterostructure". Such heterostructures consisting of, e.g. graphene and TMDs, have recently shown great promise for high-performance optoelectronic applications. For instance, the integration of graphene and monolayer WS_2 enables sensitive photodetectors with ultrahigh photoresponsivity[2], by combining strong light absorption in WS_2, efficient charge transfer across the interfaces, and the superior charge transport properties in graphene. Currently, devices are developed through trial-and-error since an in-depth understanding of interfacial charge transfer and recombination has so far remained elusive, limiting the optimization of device performance.

In this presentation, I will present our recent studies on interfacial charge carrier dynamics in graphene-WS_2 heterostructures by complementarily probing the ultrafast terahertz photoconductivity in graphene and the transient absorption (TA) dynamics in WS_2.[3] Intriguingly, THz photoconductivity studies report an extremely long-lived charge separation (beyond 1 ns) at graphene-WS_2 interfaces following optical excitations and CT. In contrast, TA studies in our study and previous reports[4-5] demonstrate a short-lived (~1 ps) charge occupation to the WS_2 exciton states following CT. The combined spectroscopic results suggest the critical role of defects on governing the interfacial CT and recombination in graphene-TMD interfaces: these defects at interfaces can efficiently "capture" (within ~ 1 ps) and "store" (over ~ 1 ns) the transferred charge carriers across the heterojunction, leading to effective long-lived photogating effect in graphene. Furthermore, for the CT process itself, we show that pump photon energies play a critical role on the CT pathways across graphene-WS_2 interfaces: when only graphene is excited (with $h\nu < E_g$, the optical bandgap energy of WS_2), it occurs via photo-thermionic emission. On the other hand, direct hole transfer from WS_2 to the valence band of graphene prevails for above-E_g excitations. Finally, we demonstrate that controlling the defect density by electrical gating can serve as an effective approach to tune the nature (e.g. the direction of the gating field) and efficiency of the photogating effect. Overall, our study not only unveils the beneficial effect of interfacial defect states, but also develops effective methods to tune their nature and density for achieving efficient photodetectors based on 2D materials and interfaces.

Reference

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