Ultrafast Charge Separation and Long-lived Photogating Effect at van der Waals Interfaces

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Integration of graphene and transition metal dichalcogenides (TMDCs) enables the assembly of van der Waals (vdW) heterostructures for optoelectronic devices by exploiting the synergy between high charge mobility in graphene and strong light-matter interactions in TMDCs [1]. At the heart of device performance are interfacial processes, namely, interfacial charge transfer and recombination. Despite recent advances in optoelectronics, the fundamental understanding of interfacial processes still lags behind. Here, we investigate these interfacial processes at photoexcited graphene-WS_2 vdW interfaces by complementarily measuring the ultrafast photoconductivity change in graphene by terahertz spectroscopy and the excited-state dynamics in WS_2 by transient absorption spectroscopy. By tuning the pump photon energy across the A-exciton resonance of WS_2, graphene or the entire heterostructure can be selectively excited. We observe a transition in CT mechanism and efficiency: sub-A-exciton excitation leads to a relatively inefficient (~1%) hot electron transfer from graphene to WS_2 via photothermionic emission, while above-A-exciton excitation results in a relatively efficient (~5%) direct hole transfer from the valence band of WS_2 to graphene [2]. Furthermore, we show that the injected or photogenerated electrons in WS_2 only occupy the excited states for ~1 ps and get trapped in defect states for over ~1 ns. This leads to a long-lived photogating effect in graphene. As such, our study unveils the bright side of defects: although typically detrimental to electrical devices, defects can be beneficial for optoelectronics. Our results elucidate interfacial processes in vdW heterostructures and suggest a potential approach, i.e., defect engineering, to further optimize the performance of optoelectronic devices.

References

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Figures

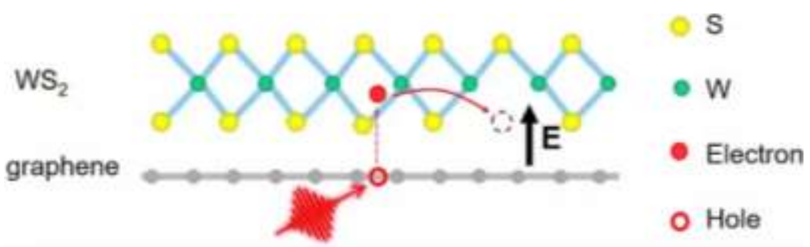

Figure 1: Defects to the rescue - following photoexcitation and interfacial CT, interfacial defects capture and store injected electrons, leading to long-lived charge separation and an interfacial electrical field (*E*) to gate graphene.