Directional ultrafast charge transfer in a $WSe_2/MoSe_2$ heterostructure selectively probed by time-resolved SHG imaging microscopy

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Heterostructures of transition metal dichalcogenides (TMD) feature a type-II band alignment which can separate photoexcited electrons and holes into different layers through ultrafast charge transfer [1] and can host long-lived interlayer excitons due to their spatially indirect nature [2]. While this charge transfer is essential for potential application, the underlying mechanisms still remain elusive [3]. Main drawbacks of previous approaches were insufficient time-resolution of the employed microscopy setups and deficiencies of linear optical spectroscopies to address individual layers of the heterostructure selectively.
Here, we introduce a new experimental concept for investigating ultrafast charge-transfer processes in TMD heterostructures by means of optical pump second-harmonic probe microscopy [4]. Our technique combines the advantages of time-resolved optical second-harmonic generation (SHG) with an optical microscopy setup. On the one hand, the method allows for pump-probe experiments in µm small structures with a superior time-resolution. On the other hand, the tensorial nature of the second-order nonlinear susceptibility allows us to distinguish the response from differently oriented layers to elucidate directional interlayer charge transfer as demonstrated for a rotationally mismatched $WSe_2/MoSe_2$ heterostructure. Thus, by combining polarization- and time-resolved measurements, very clear and systematic experiments can be performed for a variety of heterostructures to correlate observed transient changes with the underlying structure.

References

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Figures

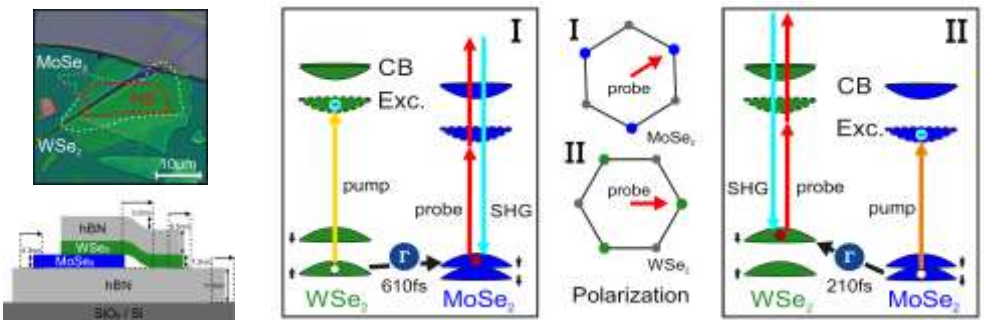

Figure 1: Directional interlayer charge transfer in a TMD heterostructure probed by time-resolved SHG