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Time-resolved second-harmonic imaging microscopy: ultrafast processes in ultrathin materials

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Internal interfaces between two solids play a decisive role in modern materials sciences and their technological applications. Among the most prominent examples are certainly semiconductor devices which have been miniaturized to such an extent that their optical and electronic properties are determined decisively by the interfaces.

Two-dimensional heterostructures of transition metal dichalcogenides (TMDCs) represent very well-defined and at the same time highly versatile model systems of van-der-Waals interfaces which enable us to investigate the electron dynamics at the interface between two molecularly thin semiconducting layers. Many combinations of two different TMDC materials feature an alignment of the electronic bands which facilitate the separation of photoexcited electrons and holes into different layers through ultrafast charge transfer leading to the formation of so-called interlayer excitons. Moreover, the electronic coupling between the layers can be modified by variation of the stacking angle, which is expected to strongly influence the exciton dynamics.

To investigate the ultrafast charge-transfer dynamics in twisted TMDC heterostructures we use time-resolved second-harmonic generation (SHG) imaging microscopy. As a nonlinear optical technique SHG is particularly suited for the investigation of interfaces and is highly sensitive to symmetries of crystalline samples. This enables us to determine the orientation of TMDC monolayers and, moreover, to probe individual layers of the twisted heterostructures by selecting the proper laser polarization. By imaging the SHG signal via a camera lens on a CCD chip, the method enables pump-probe experiments in µm small structures with a time resolution of 10 fs.

Category

Solid State (Experiment)

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