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Transient SHG microscopy on atomically thin 2D materials

Abstract

A pump-probe-based time-resolved spectroscopy has been extensively adopted to study ultrafast carrier dynamics of nanosized materials requiring sub 10 ps level time resolution. More recent research in this area has focused on two-dimensional (2D) materials. Potential applications include energy storage and the enhancement of optical susceptibility and carrier transfer efficiency. Recently, the usage of time-dependent transient second-harmonic generation (TSHG) spectroscopy was suggested for the observation of carrier dynamics and acoustic phonons with diffraction-limited spatial resolution in MoS_2 crystalline structures [1]. Compared with this work, our technique involves a simple measurement system that can deliver high pulse power density at low beam energies, whereas the use of a raster scanning system enabled the pulse exposure time to be reduced to approximately 10 μs per pixel, leading to reduced sample damage [2,3].

Specifically, we report the ultrafast carrier dynamics in atomically thin molybdenum sulphide (MoS_2) crystals, triggered by creating an A-excitonic resonance condition with pump beam. The power dependence suggests the highly dominant nature of χ_3 term in TSHG signals while the polarization dependence resolves a crossed contribution of χ_2. The thickness dependent monitoring on the target sample critically allowed us to claim the early evolution of acoustic phonons in the thicker layers. Also, interestingly enough, the dynamics of different chiral edges were resolved through spectral analysis and were supported by first principal calculation and TEM.

Theory predicts there is an additional gap in the density of states near the edge of the work function of the atomically thin ReS_2 system, which has not yet been fully identified in an experimental fashion. In this paper, we suggest one way to probe the existence of it by employing the time-resolved SHG microspectroscopy where separately-controllable high energy pump and probe pulses were accessible [4]. We also interpret the origin the ultrafast time dependent carrier dynamic behaviors of ground-state-depletion and excited-state-absorption from a viewpoint of the allowance of each electronic transition by adopting the angular momentum selection rule.

References

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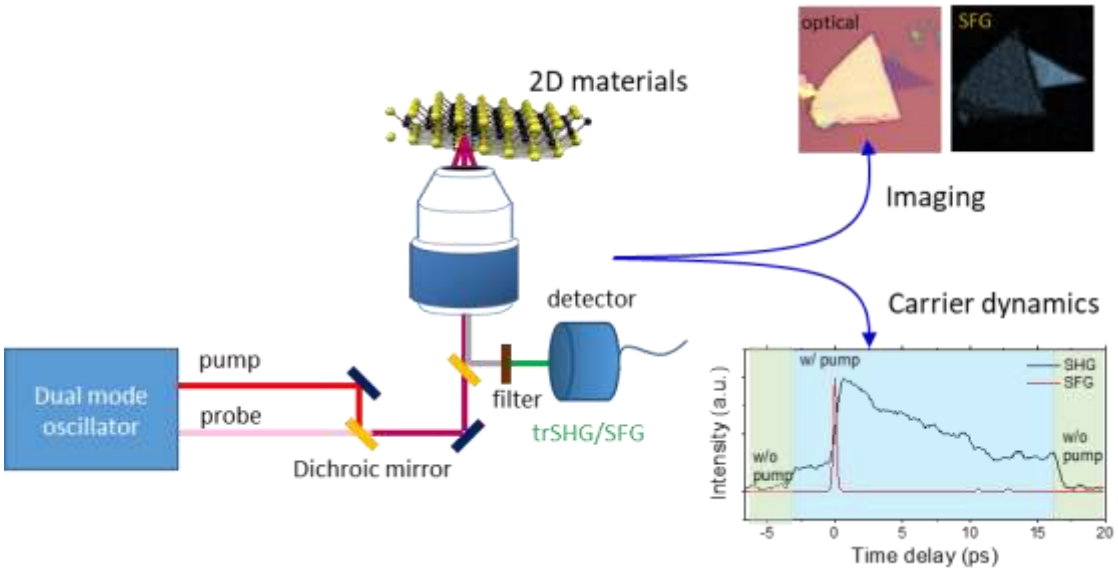

Figure 1: Scheme of time-resolved SHG microscopy on ultrafast carrier dynamics of 2D materials