Polarization-Resolved Second Harmonic Generation Imaging in 2D Materials

S. Psilodimitrakopoulos^{*}, L. Mouchliadis, G. M. Maragkakis, G. Kourmoulakis, I. Demeridou, A. Lemonis, G. Kioseoglou, E. Stratakis^{*} Institute of Electronic Structure and Laser-Foundation for Research and Technology-Hellas, GR-711 10, Heraklion, Greece

The emerging family of two-dimensional (2D) materials has provided researchers with fertile ground for exploring fundamental physical phenomena and developing innovative technological solutions. Lately, nonlinear optical measurements, including second-harmonic generation (SHG) have created new opportunities for improving the image resolution of 2D crystals [1,2] (Fig1a). At the same time, the polarization of the SHG field depends on the 2D crystal symmetry and orientation (Fig1b). Based on such SHG signal dependencies, the crystal quality of TMDs can be evaluated using polarization-resolved SHG (P-SHG) imaging [1,2] (Fig1c). Moreover, 2D TMDs can be assembled in vertical stacks. This creates new physical properties that depend on the relative orientation (twist angle) between the TMD monolayers. P-SHG imaging provides precise and real-time measurement of the twist angle, which is of utmost importance for characterizing a twisted 2D TMD heterostructure [3] (Fig1d). Additionally, degenerate minima in momentum space -valleys- in 2D materials provide an additional degree of freedom that can be used for information transport and storage. P-SHG imaging reveals that the temperature-induced changes of the P-SHG, is a unique fingerprint of valley population imbalance (VPI) [4] (Fig.1e). We envisage the optical P-SHG imaging as a powerful tool for the characterization of 2D TMD heterostructures and the engineering of their physical properties for emerging applications.



Figure 1: (a) Experimental setup for P-SHG imaging microscopy, (b) coordinates system for the theoretical model describing the P-SHG from 2D TMDs. (c) P-SHG imaging of crystal imperfections (d) Real-time imaging of twist-angle, (e) P-SHG imaging of VPI.

References

S. Psilodimitrakopoulos, L. Mouchliadis, I. Paradisanos, et al. Ultrahigh-resolution nonlinear optical imaging the armchair orientation in 2D transition metal dichalcogenides. Light Sci Appl 7, doi: 18005 10.1038/lsa.2018.5 (2018).
G. M. Maragkakis, S. Psilodimitrakopoulos, L. Mouchliadis, et al. "Imaging the crystal orientation of 2D transition metal dichalcogenides using polarization-resolved second-harmonic generation," Opto-Electron. Adv. 2, 190026, (2019).
S. Psilodimitrakopoulos, L. Mouchliadis, G. M. Maragkakis, et al. "Real-time spatially resolved determination of twist angle in transition metal dichalcogenide heterobilayers," 2D Materials, 8, 015015, (2021).

[4] L. Mouchliadis, S. Psilodimitrakopoulos, G.M. Maragkakis, et al. "Probing valley population imbalance in transition metal dichalcogenides via temperature-dependent second harmonic generation imaging," npj 2D Mater. Appl **5**, 6, (2021).

^{*} stratak@iesl.forth.gr

^{*} sopsilo@iesl.forth.gr