INSTITUT DE PHYSIQUE ET CHIMIE DES MATERIAUX DE STRASBOURG
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Séminaire AXE 1 - Sciences et Matériaux Quantiques

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Second harmonic interference & molecular excitons in 2D crystals

Optical second-harmonic generation (SHG) spectroscopy has been established as a unique probe for the structure and electronic properties of two-dimensional (2D) materials because of its symmetry-specific polarization dependence, strong excitonic resonances and the atomic thickness circumventing the phase matching requirement. When the phase information is obtained in addition to the amplitude of the SH fields, the nonlinear spectroscopy can be more powerful. In this talk, I will introduce interferometric SHG spectroscopy from principles to applications. Using spectral phase interferometry (SPI), a frequency-domain technique, we characterized the elliptically polarized SH fields generated in MoS_2/WS_2 heterobilayers [1] and photoinduced charge-transfer interactions in several types of heterobilayers [2]. Difference phase SPI allowed us to determine the material-specific SHG phase, which endows chemical sensitivity to SHG spectroscopy. Two interfering SH fields generated in heterobilayers could be comprehensively disentangled via the polarization-resolved SPI method, which enables one-shot measurements of stack angles. I will also show that SHG polarimetry can be an efficient structural probe, domain imaging and crystallinity, for hexagonal BN representing 2D dielectrics. In the second part focusing on 2D molecular crystals [3, 4], I will present our recent progress in understanding the behaviors of excitons, which undergo singlet exciton fission, superradiance, and Frenkel-charge transfer exciton mixing. These findings offer promising prospects for designing novel materials and devices with tailored photophysical properties.

References

[1] W. Kim et al., Nano Lett. **20**, 8825 (2020)
[2] W. Kim et al., ACS Nano **17**, 20580 (2023)
[3] S. Koo et al., Nano Lett. **21**, 6600 (2021)
[4] D. Kim et al., Nat. Commun. **14**, 2736 (2023)