Interferometric Second-Harmonic Generation Spectroscopy of Two-Dimensional Materials

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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, which have been or are to be reported by my group. 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 [3]. 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 [4]. I will also show that SHG polarimetry can be an efficient structural probe, domain imaging and crystallinity, for hexagonal BN representing 2D dielectrics [5]. The SH analog of Young's interference, demonstrated in these works, showcases the potential of interferometric parametric generation through atom-thick nonlinear optical materials.

References

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Figures

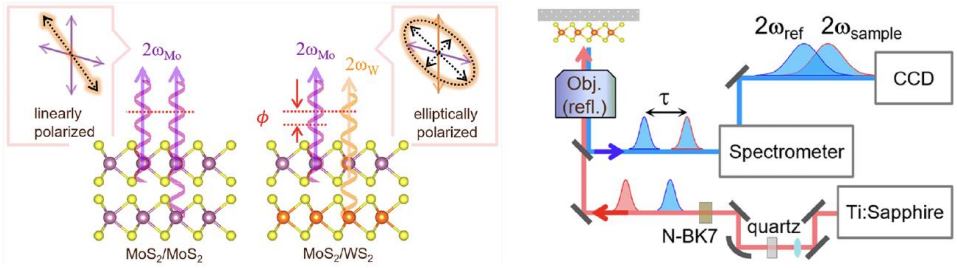

Figure 1: SH interference in bilayers (left) and interferometric SHG setup to obtain SHG phase (right)