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ABSTRACT:

From MoS₂ to NbSe₂: Understanding Nonlinear Optical Mechanisms Across 2D-LTMDs

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This talk reviews the intensity-dependent nonlinear optical (NLO) response of two-dimensional layered transition metal dichalcogenides (2D-LTMDs), spanning semiconducting (MoS₂, WS₂), semimetallic (ZrTe₂, WTe₂), and metallic systems (NbS₂, NbSe₂). Focusing on liquid-phase suspensions obtained via redox exfoliation, we discuss how distinct nonlinear mechanisms emerge across this material family and how they are strongly governed by the spectro-temporal excitation regime. Using a combination of complementary techniques—including hyper-Rayleigh scattering, Z-scan, photoacoustic Z-scan, optical Kerr gate, spatial self-phase modulation, and Fourier-transform nonlinear optics—we disentangle thermal and electronic contributions to the nonlinear response. Particular attention is given to the third-order susceptibility, where both nonlinear refraction and absorption exhibit rich, intensity-dependent behavior. For instance, metallic NbS₂ displays a remarkable sign reversal of the nonlinear refractive index with increasing intensity, while semiconducting MoS₂ reveals higher-order (fifth-order) contributions under femtosecond excitation.

A central theme of the talk is the clear distinction between slow thermal nonlinearities—dominant in high-repetition-rate or continuous-wave regimes—and ultrafast electronic responses observed under femtosecond excitation. Thermal effects, often mediated by solvent heating in suspensions, can lead to large effective nonlinearities, whereas intrinsic electronic contributions are faster but significantly smaller in magnitude.

By systematically comparing materials from MoS₂ to NbSe₂, this work provides a unified framework linking electronic structure, absorption properties, and nonlinear optical mechanisms. The results highlight both fundamental insights and practical considerations for exploiting 2D-LTMDs in photonic and optoelectronic applications, emphasizing the importance of correctly identifying the physical origin of the observed nonlinearity.