Extreme nonlinear optics in redox-exfoliated MoS_2

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Transition metal dichalcogenides have been noted for their nonlinear optical properties, including large second harmonic generation, saturable absorption, and high harmonic generation. In this talk, I will describe high-order multiphoton photoluminescence and saturation of second harmonic generation in multilayer MoS_2 prepared by redox-exfoliation. The mechanisms of nonlinear optical emission from MoS_2 were studied with Fourier-transform nonlinear optical spectroscopy (FT-NLO), which yields a nonlinear excitation spectrum containing information about the photon order of the nonlinearity as well as multi-photon resonances. Analysis of the FT-NLO spectra of MoS_2 revealed high-order multiphoton photoluminescence (up to 10^{th} order), which is obscured in power-dependent measurements, which only report on the average photon order. Unexpected high-order nonlinearities were also observed in coherent second harmonic generation, and determined to result from saturation of the two-photon exciton resonance which is known to enhance second harmonic generation in MoS_2. Comparison to MoS_2 prepared by MOCVD demonstrates that these high order nonlinearities are unique to redox-exfoliated MoS_2, suggesting avenues for tailoring the nonlinear optical properties of 2D semiconductors with redox chemistry.