Unraveling fundamental processes in two-dimensional nanomaterials with ultrafast nonlinear optical microscopy

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Ultrafast optical microscopy (UOM) has recently emerged as a powerful tool for studying the properties of nanoscale materials with femtosecond temporal resolution and micron spatial resolution. The extension of UOM beyond typical pump-probe techniques to nanoscale nonlinear optics (e.g., second harmonic generation (SHG) and magneto-optical Kerr effects (MOKE)) has provided even more insight into a wide range of materials, from nanoscale magnets and metallic nanoparticles to two-dimensional (2D) semiconductor heterostructures.

Here, I will discuss our recent measurements using UOM, especially in combination with nonlinear optics, to study a range of nanoscale semiconductors. I will begin by describing our use of time-resolved MOKE microscopy to resolve spin-phonon coupling dynamics in the 2D vdW ferromagnet CrI3. I will then discuss the use of both static and time-resolved SHG microscopy to reveal extremely large nonlinearities in the polar semiconductor BiTeBr. Finally, we have recently combined WSe_2 monolayers with a dielectric metasurface to not only enhance exciton emission, but also control exciton dynamics. Overall, this combination of measurements at both nanometer distance and femtosecond time scales can reveal the intrinsic properties of these 2D nanosystems, with important implications for a wide range of applications.