Probing Defect-Driven Exciton and Spin Dynamics in 2D TMDs via PL and ODMR Spectroscopy

Yusuf Selim Ocak[1], Musa Salami[1], Peker Milas[1], Pranish Shrestha[1], Sergiy Krylyuk[2], Albert Davydov[2], Chen Chen[3], Shalini Kumari[3], Nicholas Trainor[3], Abdallah El Damery[4], Can Ataca[4], Berend T. Jonker[5], Michael Spencer[6,7], and Birol Ozturk[1,7]

[1]Department of Physics and Engineering Physics, Morgan State University, Baltimore, MD
[2] National Institute of Standards and Technology, Gaithersburg, MD
[3]Materials Research Institute, Penn State University, University Park, PA
[4]Department of Physics, University of Maryland-Baltimore County, Baltimore, MD
[5]Materials Science & Technology Division, Naval Research Laboratory, Washington, DC
[6]Electrical and Computer Engineering, Morgan State University, Baltimore, MD
[7]Microelectronic Education and Research Center, Morgan State University, Baltimore, MD

Abstract:

Defects and strain-induced modifications in two-dimensional transition metal dichalcogenides (2D TMDs) significantly influence both excitonic and spin-related phenomena, presenting opportunities for tunable optoelectronic and quantum functionalities. In this study, we investigate exciton and spin dynamics in WSe_2 monolayers and GaSe multilayers through a combination of photoluminescence (PL) spectroscopy, atomic force microscopy (AFM)-based strain engineering, and optically detected magnetic resonance (ODMR).

WSe_2 monolayers synthesized via mechanical exfoliation and chemical vapor deposition (CVD) methods were characterized at cryogenic temperatures and with varying excitation laser powers. Global fitting of PL spectra revealed sharper excitonic peaks and enhanced thermal stability in CVD-grown samples, while exfoliated ones exhibited broader linewidths and greater sensitivity to external perturbations. For GaSe, thickness-dependent PL measurements showed strong blue shifts and reduced emission intensities in thinner flakes, consistent with quantum confinement and interlayer decoupling.

To explore localized strain effects, we applied nanoindentation using AFM on WSe_2 monolayers. The resulting PL spectra exhibited pronounced shifts in excitonic peak positions, linewidth broadening, and altered power-law scaling, indicating modified recombination pathways and exciton-phonon interactions in strained regions.

ODMR spectroscopy, combined with DFT-based calculations, was employed to probe the spin characteristics of defect states in WSe_2 monolayers. Theoretical modeling of WSe_2 structures predicted zero-field splitting (ZFS) for point and edge defects, including zigzag and armchair configuration edge defects. Experimentally, we detected ODMR dips within the 1-2 GHz range under cryogenic conditions for nano-indented WSe_2 monolayers, aligning well with the predicted ZFS values.

This integrated approach provides new insights into the interplay of defects, strain, and spin-resonant excitons in 2D TMDs, supporting their potential use in quantum sensing and optoelectronic applications.