Optical Probing of Magnetic Ordering in van der Waals Materials

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The discovery of long-range magnetism in two-dimensional van der Waals (vdW) materials has opened new opportunities to explore magnetic phenomena in reduced dimensionality and to integrate magnetic order into layered heterostructures (HSs). The interplay between sample thickness, external magnetic fields, and optical excitations governs the stabilization of magnetic ordering, including the formation of topological spin textures. These advances substantially broaden the scope of spintronic, valleytronic, and quantum magneto-optical applications.

In the first part of this talk, I will explore the temperature dependence of spin-phonon coupling in three magnetic vdW materials with Cr atoms as the source of magnetic ordering, *i.e.* $Cr_2Ge_2Te_6$,[1] $CrBr_3$,[2] and $CrCl_3$,[3] using Raman scattering (RS) spectroscopy. $Cr_2Ge_2Te_6$ and $CrBr_3$ exhibit ferromagnetic (FM) intralayer and interlayer coupling and strong out-of-plane magnetic anisotropy, resulting in spins aligned perpendicular to the layers. In contrast, $CrCl_3$ displays FM intralayer order with an in-plane easy axis and antiferromagnetic (AFM) interlayer coupling. Symmetry analysis, supported by first-principles phonon calculations, enables the unambiguous assignment of all Raman-active modes in all three materials. Temperature-dependent RS studies reveal pronounced signatures of spin-phonon coupling across the transition from fully FM or AFM phases, through an intermediate regime characterized by local, domain-like FM order, to the paramagnetic phase.

In the second part of the talk, I will demonstrate a pathway to exploit magnetic proximity fields to activate properties in materials that would otherwise require external stimuli. This concept is realized by creating HSs comprising a bulk $CrCl_3$ antiferromagnet and a monolayer (ML) WSe_2 semiconductor.[4] Photoluminescence and magnetic force microscopy measurements reveal strong interlayer interactions in the $WSe_2/CrCl_3$ HSs. It is demonstrated that, at specific spatial locations, the magnetic proximity effect between the WSe_2 ML and the $CrCl_3$ bulk activates dark exciton emission in the WSe_2 ML. Notably, the dark exciton emission persists to temperatures higher than the intraplane Curie temperature (T_C) of $CrCl_3$, attributed to an enhanced T_C in locally strained regions of the $CrCl_3$ layer.

References:

[1] G. Krasucki et al., arXiv:2510.01881 (2025).
[2] Ł. Kipczak et al., Scientific Reports 14:7484 (2024).
[3] Ł. Kipczak et al., arXiv:2601.16927 (2025).
[4] Ł. Kipczak et al., Nanoscale Horizons 10, 2465 (2025).