Magneto-Excitonic Properties From Monolayer to Trilayer CrSBr

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Layered magnetic materials (LMMs) offer a versatile platform for exploring the interplay between reduced dimensionality, magnetism, and excitonic phenomena. Properties that are highly desirable for modern opto-, spin-, and valleytronics applications [1]. Here, we investigate the magneto-excitonic properties of the anisotropic van der Waals antiferromagnet CrSBr from the monolayer to trilayer limit. Low-temperature reflectance contrast, photoluminescence (PL), and photoluminescence excitation (PLE) spectroscopy are employed to resolve multiple layer-dependent excitonic transitions and to examine their distinct responses to an external out-of-plane magnetic field ($\vec{B}||\hat{c}$).

The layer-resolved PL spectra exhibit two emission features, labeled A and A' at around 1.3 eV [2]. The corresponding PLE spectra reveal two higher-energy excitonic resonances, denoted B and C, at approx. 1.8 eV and 2.2 eV, respectively. Upon application of an $\vec{B}$, spin canting induces a moderated red-shift of about 10 meV in the A/A' and C transitions. In contrast, the B resonance experiences a red-shift nearly 10 times larger ($\sim$100 eV). This pronounced behavior reflects magnetic-field-induced modifications of the electronic band structure [3, 4], particularly affecting the electronic states involved in the B transition.

Our magneto-PLE measurements demonstrate that the distinctive excitonic duplexity observed in CrSBr, supporting both Frenkel- and Wannier-Mott-like excitons [4], persists even in the atomically thin limit. These results highlight the robustness of magneto-excitonic coupling in CrSBr, challenging the conventional dichotomy between Frenkel and Wannier excitons, and opening new opportunities for exploring hybrid excitonic phenomena in 2D materials.

References:

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