Magneto-Optical Layer Dependence of CrSBr

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Two-dimensional (2D) layered magnetic materials (LMMs), such as CrI_3, $FePS_3$, and CrSBr, form a newly emerging class of van der Waals (vdW) materials highly suitable for modern opto-, spin-, and valleytronics devices. Due to the magneto-optical coupling, magnetic transitions can be observed by optical measurements. In this work, we investigate CrSBr thickness dependence through reflectance contrast (RC), photoluminescence (PL), and Raman scattering (RS), mainly as a function of temperature.

Considering the bulk CrSBr, temperature evolution of the PL spectra in the range from T=5 K up to 300 K is shown as a false color map in Fig. a. A complex PL spectrum is observe at low-temperature, close to E=1.36 eV. The bunch of emissions lines are related to excitons and exciton-polaritons. [1] The temperature rising reveals two optical transitions at T=70 K and 140 K. While the last one occurs to the expected Néel temperature (T_N), marking the ending of the antiferromagnetic (AFM) ordering, the first one is related to fluctuations of the magnetic order. [2] Meanwhile, the 8 layers (L) (approx. 6.7 nm) CrSBr, low-temperature PL spectrum present a much simple lineshape (Fig. b). Only one emission line at approx. E=1.36 eV is appreciate, which is followed by a low-energy broad band. As further as the temperature increases the emission line slightly redshifted. However, an inflection is observed at T=70 K, and the line goes towards the broad band faster. The PL line and the broad band merged at T_N=140 K.

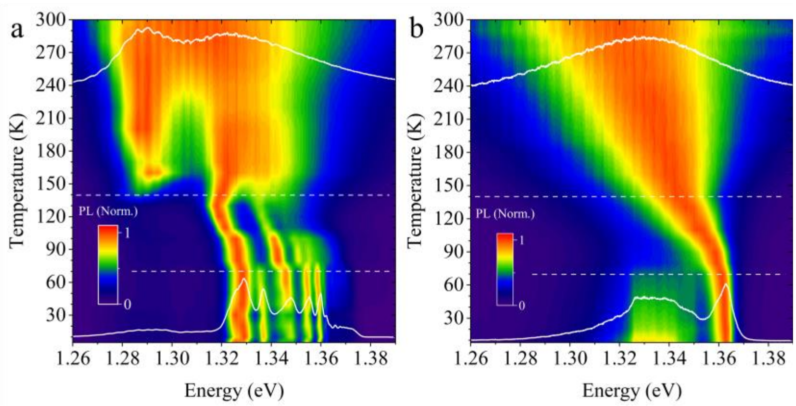

Figure: The temperature evolution of the PL spectra in CrSBr bulk (a) and 8 L (b). The measurements were taken under λ=515 nm excitation.

Our results confirmed a strong magneto-optical coupling. The study of thickness dependence of LMM can provide us more details about the magnet phases, as well as the coupling exciton-vibration due to the magnetism. These findings contribute to our understanding of the fundamental characteristics of LMM and their potential applications, yielding additional insights into its unique properties.

[1] Wilson, N.P., Lee, K., Cenker, J. *et al.*, *Nat. Mater.* **20**, 1657–1662 (2021).
[2] Kaiman Lin, Xiaoxiao Sun *et al.*, ACS Nano 2024 18 (4), 2898-2905