Using excitons to investigate magnetic phases in CrSBr analogs

S. Badola^{1, *}, A. Pawbake¹, K. Mosina², A. Soll², Z. Sofer², C. Faugeras¹

¹Laboratoire National des Champs Magnetiques Intenses, CNRS-UGA-UPS-INSA-EMFL, 38042, Grenoble, France ²Department of Inorganic Chemistry, University of Chemistry and Technology Prague, Technicka 5, 166 28 Prague 6, Czech Republic

Elementary excitations like exciton are one of the representative of light matter interactions and are of prime importance in condensed matter physics to realize opto-electronic applications. An additional degree of freedom is granted when excitons are hosted by a magnetic crystal and that the electronic band structrue is related to the magnetic ground state providing a tuning knob to either control optical properties by modifying magnetism or to access and possibly modify the magnetic properties by adressing excitons. In this regard, the A-type antiferromagnetic van der Waals semiconductor CrSBr appears today as unique as it displays exceptional coupling between optical (excitonic) and magnetic properties [1, 2, 3]. Magneto-crystalline anisotropies in CrSBr are more important than the interlayer exchange interaction and previous studies have demonstrated that bulk CrSBr is very sensitive to strain [4] or pressure [5] that can be used to drive CrSBr into the weak anisotropy regime.

Today, bulk CrSBr appears as a unique system showing such an intimate coupling between its magnetic and optical properties. Here, we consider alloys of CrSBr with either Chloride atoms to replace Bromide $(CrSBr_xCl_{(1-x)})$ or with Vanadium in substitution with the magnetic Chromium (CrSBr : V) atoms and we explore the magneto-optical properties of this family of materials using magneto-photoluminescence and reflectance measurements. We show that in the space of explored compositions, these materials are direct band gap semiconductors hosting excitons. Similar to CrSBr, these alloys also display a coupling between the magnetic ground state and the electronic band structure making it possible to address the magnetic ground state by generating excitons. We show that the values of critical magnetic fields necessary to saturate the spin lattice decrease when alloying chloride or vanadium and that alloying also affects the magneto-crystalline anisotropies, driving for some compositions the alloy into the weak anisotropy regime where a field induced spin-flop magnetic phase can be observed.



Figure 1: Representative low temperature photoluminescence spectra of $CrSBr_xCl_{(1-x)}$ with x = 1,0.8,0.5 and of CrSBr : V.

References

- [1] N. Wilson et al. Nat. Mater. 20, 1657 (2021).
- [2] E Telford, et al. Adv. Mater. 32, 2003240 (2020).
- [3] K Lin, et al. ACS Nano 18,4, 2898 (2024).
- [4] J. Cenker et al. Nat. Nanotech 17, 256 (2022).
- [5] A Pawbake, et al. Nano Letters 23, 9587-9593 (2023).

^{*}E-mail: shalini.badola@lncmi.cnrs.fr