High Energy Magnetic Excitons In CrSBr

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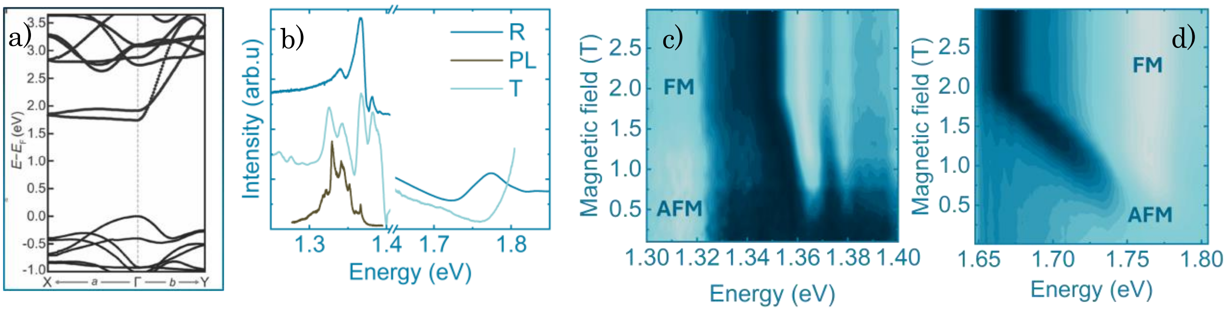
Chromium Sulfide Bromide (CrSBr) is a layered material that has very recently gained significant interest due to its unique combination of magnetic and semiconducting properties and excellent chemical stability in ambient conditions.[1] In its pristine form CrSBr exhibits a layered structure with strong in-plane magnetic ordering below Néel temperature. Each layer couples antiferromagnetically to the neighbouring layers. This antiferromagnetic state can be overcome using relatively low external magnetic fields applied parallel to one of the three magnetization axes, affecting electronic structure significantly.[3]

This study focuses on the excitonic properties of CrSBr in its ferromagnetic phase, under the influence of extreme magnetic fields, up to 67T parallel to the hard magnetization c axis. We present preliminary results of the study over a higher excitonic species coming presumably from a split valence band at Γ point in the Brillouin zone (fig. a,b). This higher energy exciton (1.7 eV) is strongly coupled to magnetic order and shifts by significant 93 meV between the anti-ferromagnetic and ferromagnetic phase . This is around the order of magnitude stronger change than the one characterising fundamental excitonic transition at 1.3eV, (11 meV shift) which can be seen in (fig.c and d). This study is expanded by the investigation of the behaviour of these excitonic transitions in the FM phase, up to 65T, where high energy exciton experiences weak diamagnetic shift of ~0.2μeV/T^2 . In contrast to ground exciton state which shift in high magnetic field is not detectible. We ascribe this observation to different nature of two exciton states where the lower energy one is of Frenkel type while the higher energy exciton is of Wanier type with wave function expanding over neighbouring layers. Additionally, we investigate the temperature dependence of the optical response in high magnetic fields for both a thin (~10nm) layer of CrSBr transferred to an optical fiber and a bulk crystal to understand how the competition between magnetic order and thermal disorder affects the excitonic properties of this material.



[1] Lee, Kihong, et al. "Magnetic order and symmetry in the 2D semiconductor CrSBr." Nano Letters 21.8 (2021).

[2] Ziebel, Michael E., et al. "CrSBr: An Air-Stable, Two-Dimensional Magnetic Semiconductor." Nano Letters (2024).

[3] Wilson, N.P., Lee, K., Cenker, J. et al. Interlayer electronic coupling on demand in a 2D magnetic semiconductor. Nat. Mater. 20, 1657–1662 (2021)