Dielectric tensor, magnetic anisotropies and coupled excitations in layered magnetic semiconductors CrSBr

P. M. Piel, T. Kliewer, J.-H. Larusch, S. Schaper, A. Łopion, <u>U. Wurstbauer</u>

Institute of Physics and Center for Soft Nanoscience (SoN), University of Münster, Germany

.

Two-dimensional materials exhibit unique properties due to their atomically thin structure and weak van der Waals (vdW) coupling between layers resulting in layer dependent properties. As in the case of the layered magnetic semiconductor CrSBr, individual layers are ferromagnetically ordered below the Neel temperature (T_{N≈132K}), while adjacent layers are coupled antiferromagnetically. Due to the highly anisotropic electronic bands in CrSBr, electronic and excitonic states at the fundamental band-gap behave quasi-one-dimensional [1]. Moreover, the resulting excitonic transitions are highly sensitive to the collective spin order. Below the critical temperature, an external magnetic field applied along the magnetic hard directions drives the system from the antiferromagnetic into a

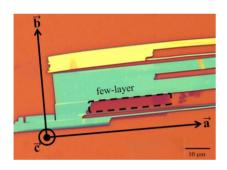


Fig.1. Optical micrograph of a CrSBr bulk flake with a few-layer region marked. Triaxial anisotropy of the crystal lattice impacts electronic, optical and magnetic properties of the material.

ferromagnetically ordered state causing a quadratic red-shift of the exciton energies theoretically explained by spin-allowed charge transfer changing the composition and nature of excitons [2]. By a combination of magneto-reflectance, magneto-photoluminescence and magneto resonant inelastic light scattering (RILS) experiments, we study strong coupling between charge, lattice and spin degrees of freedom as well as their changes when interfaced to other 2D magnetic semiconductors from the transition metal phosphor trisulfide group with different magnetic anisotropies.

The strong light matter interaction in thin CrSBr film is highly tunable by layer number and magnetic polarization. To develop a better understanding, we

access the materials dielectric tensor in the paramagnetic and ferromagnetic phase by variable-temperature spectroscopic imaging ellipsometry. In agreement with theory, we extract highly anisotropic dielectric functions along the crystallographic main axes with strong excitonic resonances particularly in the plane [4].

We acknowledge the fruitful collaboration with Florian Dirnberger, Julian Klein, Zdeněk Sofer, Marie-Christin Heißenbüttel, Thorsten Deilmann and Michael Rohlfing.

- [1] J. Klein et al. ACS Nano, 17, 6, 5316-5328 (2023).
- [2] M.-C. Heißenbüttel et al. 111, 075107 (2025).
- [3] F. Dirnberger et al. Nature 620, 533–537 (2023).
- [4] P.M. Piel, S. Schaper, et al (2025).

⁺ Author for correspondence: wurstbauer@uni-muenster.de