

Dielectric tensor and coupled excitations in layered (magnetic) semiconductors

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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 ($T_N \approx 132\text{K}$), 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 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 resonant inelastic light scattering (RILS) experiments in resonance with those excitons, we reveal and study strong coupling between charge, lattice and spin degrees of freedom.

Moreover, thin CrSBr film host self-hybridized polaritons [3]. To develop a better understanding of the extraordinary light-matter interaction in CrSBr, we access the materials dielectric tensor in the paramagnetic phase by spectroscopic imaging ellipsometry that is hard to access by alternative approaches such as reflectance measurements due to the strong anisotropy. In agreement with theory, we extract highly anisotropic dielectric functions along the crystallographic main axes with strong excitonic resonances particularly in the plane [4].

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[2] M.-C. Heißenbüttel et al. arXiv:2403.20174.

[3] F. Dirnberger et al. Nature 620, 533–537 (2023).

[4] P.M. Piel, S. Schaper et al (2024).