# Excitons in 2D Magnets

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NREL/PO-5F00-91308

*Theoretical abilities that we built*: In our QS$G\hat{W}$ formalism[1], the electronic eigenfunctions are calculated self-consistently in presence of the ladder $e$-$h$ vertex corrections to the screened Coulomb exchange. Charge and self-energies are recalculated iterated until the desired tolerance is achieved in the one-particle Green's function. Further, to include the multi-determinantal (spin-flip structure of multiplets) nature of many body correlations we combine QS$G\hat{W}$ with DMFT. Below we explore various classes of strongly correlated magnetic systems where excitons are sufficiently described within either the QS$G\hat{W}$ or the QS$G\hat{W}$+DMFT approach. In the process, we establish the digrammatic requirements for a minimum-sufficient theory for describing excitons in large classes of materials.

## Excitons in 2D ferromagnets

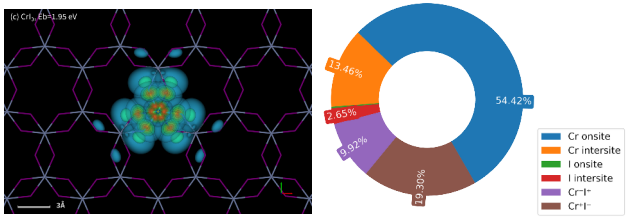
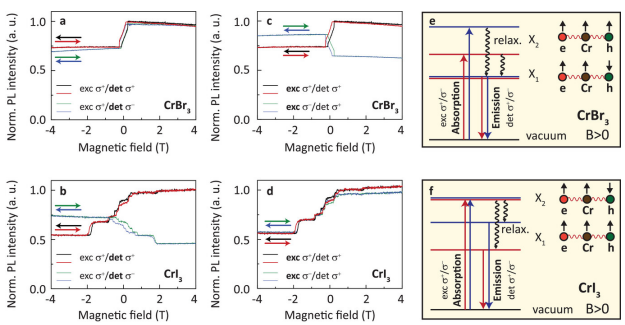



Figure 1. The ground state excitonic wavefunction in $CrI_3$ is shown in real space and its e-h probability density is decomposed in different atomic channels.

If the excitons for different X (X=Cl, Br, I) derive mostly from atomic Cr, they should be similar and they should interact with an external magnetic field in a similar fashion. However, that is not the case. We show that with combined theory and experimental work[4].




| | Ground excitonic state | | Excited excitonic state | |
|---|---|---|---|---|
| | Cr-electron | Cr-hole | Cr-electron | Cr-hole |
| $CrBr_3$ | FM | AFM | FM | FM |
| $CrI_3$ | FM | FM (small exchange constant) | FM | AFM |

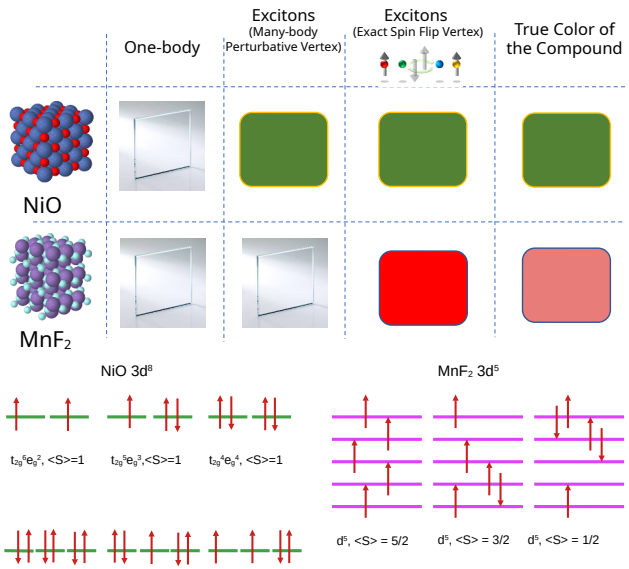
Figure 2. The magneto-photo PL is studied for ground (a,b) and excited (c,d) state excitons. Spins of the electrons and holes (that form excitons) couple differently with magnetic field (shown in a summary in (e,f) and in the table).

### Scientific Achievement[1]

**Quantitative distinctions between onsite 'forbidden' $d$-$d$ and intersite 'allowed' $d$-$d$ and $p$-$d$ components**: all excitons are captured within QS$G\hat{W}$ theory; some are dark, some are bright. The theory enables a quantitative understanding of their emergence and degree of brightness without subscribing to any Hubbard or extended Hubbard Hamiltonian.

**Beyond Ligand-field theory**: QS$G\hat{W}$ explains quantitatively the nature and extent of onsite and intersite $d$-$d$, $p$-$d$ components determines how spins couple differently in different materials and also for ground and excited states.

## Excitons in 3D antiferromagnets: a theory for colors




### Scientific Achievement[2]

**Excitonic absorption that determines the Green color of NiO** is captured within many-body perturbative approach: because it does not involve the spin flip component of the vertex function. QS$G\hat{W}$ theory is sufficient for that.

**Excitonic absorption that determines the pink color of $MnF_2$ is not captured within many-body perturbative approach but in DMFT:** since such absorptions derive from a transitions between majority spin and minority spin d states on Mn, and hence, a spin flip process that also modifies the optical response is required – this high order process not contained in the usual perturbation theory but is contained within DMFT[5].

**The body of literature loosely attributes these excitons to symmetry lowering mechanisms such as spin-orbit coupling, coupling with odd-parity phonon modes and so forth:** and we show that such analysis is wrong and we quantitatively demonstrate that the diagrammatic mechanism that lead to the emergence of these excitons and their brightening mechanisms are two completely different aspects which should not be confused.

**Bottom line: unless spin-flip processes are involved, we believe, QS$G\hat{W}$ is a sufficient theory for describing excitons, irrespective of their degree of wavefunction localization**.

[1] B. Cunningham et al., *QS$G\hat{W}$: Quasiparticle Self consistent $GW$ with ladder diagrams in $W$*, Phys. Rev. B 108, 165104 (2023).

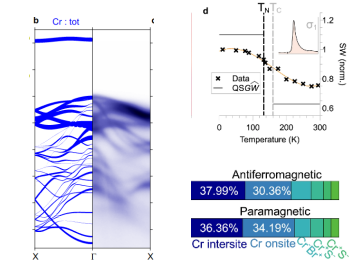
[2] S. Acharya et al, *Electronic structure of chromium trihalides beyond density functional theory*, Phys. Rev. B, 104, 155109 (2021).

[3] S. Acharya et al, *Real and momentum space description of the excitons in bulk and monolayer chromium tri-halides*, npj 2D Materials and Applications, 6, 33 (2022).

[4] M. Grzeszczyk et al., *Strongly Correlated Exciton-Magnetization System for Optical Spin Pumping in $CrBr_3$ and $CrI_3$*, Adv. Mater. 35, 2209513 (2023).

## Impact of magnetic disorder on excitons in low dimensional magnets

We explore the impact of magnetic order-disorder transitions on exciton's oscillator strengths. The figures show one- and two- particle properties of CrSBr.

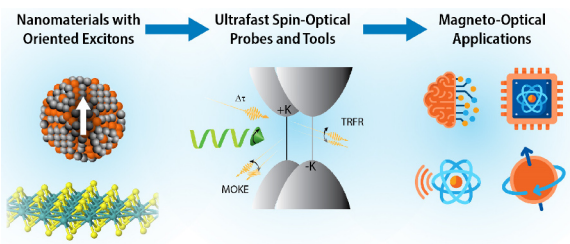



### Scientific Achievement[3]

**ARPES** We compare QS$G\hat{W}$ electronic structure against high quality ARPES data for both AFM and PM phases, showing the remarkable fidelity of the theory [6,7]. This work also solves an outstanding problem regarding the band gap values and exciton binding energies for CrSBr.

**Excitons** become moderately darker as the system magnetically disorders [7]. The disordering of the FM chains in the PM phase hinders the electron-hole hopping leading to more localized excitons with larger proportions of onsite $d$–$d$ components and lesser oscillator strength.

## Review paper




### Scientific Achievement[4]

**This review** [8] discusses several outstanding issues on 2D magnets, applications of solid-state and quantum chemical approaches; where they meet and diverge, and future roadmap on applications of vdW magnets in quantum-nanotechnology, and spin-photonics.

[5] S. Acharya et al., *A Theory for Colors of Strongly Correlated Electronic Systems*, Nature Communications 14, 5565 (2023).

[6] M. D. Watson, et al, *Giant exchange splitting in the electronic structure of A-type 2D antiferromagnet CrSBr*, Accepted in npj 2D Mater. and Applications. M Bianchi et al., *Paramagnetic electronic structure of CrSBr: Comparison between ab initio GW theory and angle-resolved photoemission spectroscopy*, Phys. Rev. B 107, 235107 (2023).

[7] F. Ruta et al, *Hyperbolic exciton polaritons in a van der Waals magnet*, Nature Communications 14, 8261 (2023).

[8] D. Lubert-Perquel et al, *Optically Addressing Exciton Spin and Pseudospin in Nanomaterials for Spintronics Applications*, ACS Applied Optical Materials, 1, 1742 (2023).