# Magnetooptical properties of two-dimensional magnetic semiconductors and their heterostructures

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The exploration of two-dimensional magnetic semiconductors (2DMSs) has shown great promise and interest in tuning the magnetic and electronic properties as well as studying magneto-optical effects. Recent studies show that the ordering of atomic magnetic moments in 2DMSs can be tuned by external factors and leads to significant changes in their optical spectra. We employ Density Functional Theory (DFT) and DFT-based Bethe-Salpeter Equation (BSE) to theoretically investigate the magnetooptical properties of 2DMSs, their alloys and heterostructures, with regard to their experimental spectra.

We investigate the impact of the spin direction on the optoelectronic properties of transition metal phosphorus trichalcogenides ($MPX_3$, M = Mn, Ni, Fe; X = S, Se) exhibiting various antiferromagnetic (AFM) arrangements within the 2D limit. Our analysis reveals large exciton binding energies (up to 1.1 eV), exceeding the values of transition metal dichalcogenides (TMDs). We determine the optically active band-edge transitions, revealing that they are sensitive to in-plane magnetic order. We predict the anisotropic effective masses and the type of linear polarization as important fingerprints for sensing the type of magnetic arrangements. We identify the spin-orientation-dependent features such as the valley splitting, the effective mass of holes, and the exciton binding energy. In particular, we demonstrate that for X=S, Se, a pair of nonequivalent K+ and K− points exists yielding the valley splittings that strongly depend on the direction of AFM aligned spins. Notably, for the out-of-plane direction of spins, two distinct peaks are expected to be visible below the absorption onset, whereas one peak should emerge for the in-plane configuration of spins. We propose a strategy for how the spin valley polarization can be realized in 2D AFM within a honeycomb lattice [1].

Within DFT+U approach we qualitatively explain the origin and the position of the experimentally observed mid band-gap states in layered $MnPS_3$, and corresponding peaks visible in the alloyed systems $Mn_xZn_{1-x}PS_3$. Accordingly, emission at 1.3 eV in all alloyed compounds results from recombination from a $^4T_{1g}$ Mn(II) excited state to a hybrid p-d state at the valence band, as turns out from the analysis of calculated projected densities of states [2].

We theoretically address the recent observation of remarkably strong magneto-excitonic effects in CrSBr, which exhibits optical transitions above the fundamental exciton energy with a colossal spectral shift approaching 100 meV under applied magnetic fields - an order of magnitude larger than previously observed magneto-excitonic responses. DFT-based BSE calculations quantitatively explain the microscopic origin of the observed red- and blueshifts [3].

Motivated by recent optical measurements in magnetic field, we study a $NiPS_3/WSe_2$ heterostructure with various magnetic orderings and stackings. Our DFT+U calculations reveal the presence of interlayer excitons coupled with in-gap 3d states of Ni, as well as an exotic spin texture effecting from Rashba spin-orbit coupling and exchange interactions [4].

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## References

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