Linear dichroim of magneto-photoluminescence from $CrCl_3$

B.Kardynał[1,2] , L. Zhou[1,2], M. Lezaic[1], R. Rani[1], M. Bui[1,2]

[1]Peter Grünberg Institute, Forschungszentrum Jülich, Jülich, Germany
[2]Departement of Physics, RWTH Aachen, Aachen, Germany

Chromium halides have been widely investigated as van der Waals insulators that exhibit magnetic ordering at low temperatures. Ferromagnetic ordering within individual monolayers results from the magnetic moments of chromium atoms, each of which is occupied with three electrons in the high-spin ground state of 3d orbitals, split by the crystal field into low energy triplet and high energy doublet states. $CrCl_3$ is unique among the halides in that the spins of Cr^{3+} 3d electrons are oriented in the plane of the monolayer. In this contribution, we discuss the modification of the spin orientation in the excited state of the Cr^{3+} that is revealed in the linear dichroism of magneto-photoluminescence with a magnetic field applied in the plane of the $CrCl_3$ films. In strain-free samples, the dominating photoluminescence polarisation axis is aligned with the expected orientation of spins at a given magnetic field, while lattice strain introduces linear dichroism even without an applied magnetic field. We further simulated CrCl3 in the excited state using DFT calculations using carefully prepared model system. The simulations revealed a reorientation of the excited-electron spin, which gains an out-of-plane component and which is coupled to the excitation-induced orbital moment, the latter being responsible for the linear dichroism. Finally, we comment on possible magnetic excitations that can arise from the spin reorientation under optical exciation.