## Magneto-Optical Studies of Layered Antiferromagnet CrPS<sub>4</sub>

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Two-dimensional magnetic materials have recently attracted significant attention as promising platforms for studying spin-based devices such as spin filters or valves and investigating exotic spin-dependent phenomena such as the realization of magnetic skyrmions. However, micrometer-scale dimensions of these materials make their direct magnetic characterization challenging. Conventional bulk measurement techniques are often insufficient, therefore, approaches through transport measurements or optical probes must be employed. In this work, we investigate thin-layers of antiferromagnetic semiconductor CrPS<sub>4</sub>, which is an A-type antiferromagnet in its bulk form, and explore its spin properties through magneto-optical effects and polarization-resolved photoluminescence (PL) measurements.

The few-layer flakes of CrPS<sub>4</sub> investigated in our work are obtained by mechanical exfoliation from bulk crystals which are subsequently capped with hBN. For probing the spin polarization, we measured the degree of circular polarization of PL emission from CrPS<sub>4</sub> in Faraday geometry and confirm the out-of-plane spin orientation which is further supported by the magnetic circular dichroism (MCD) measurements. We further extend our study to

the Voigt geometry, performing both MCD and circularly polarized PL measurements to track the evolution of spin orientation under in-plane magnetic fields. Taken together, these optical spectroscopic results demonstrate the potential of two-dimensional antiferromagnets as versatile exploring platforms for spin physics in van der Waals platforms and advancing next-generation spin-based technologies.

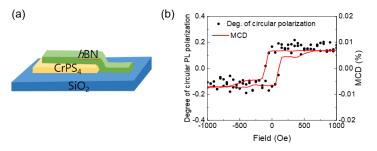


Figure 1. (a) Schematic of *h*BN capped few-layer CrPS<sub>4</sub>. (b) Degree of circular PL polarization (black dots) and magnetic circular dichroism (MCD, red line) measured as a function of magnetic field. The degree of circular PL polarization is defined as (left circular PL – right circular PL) / (left circular PL + right circular PL).

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