# Magnetic Susceptibility Curve of $CrPS_4$ from Raman Magnetospectroscopy

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2D van der Waals transition metal phosphorus chalcogenides (TMPCs) incorporating transition metal atoms such as Mn, Fe, or Cr combine advantageous properties of two-dimensional semiconductors and intrinsically antiferromagnetic materials. So far, a majority of experimental research on this appealing class of semiconductors has employed such methods as magnetometry, neutron diffraction or electrical transport. As a result, despite an increasing number of studies, a link between their magnetic and magnetooptical properties has not yet been established.

In the present work, we perform a comprehensive study on a representative member of the TMPC family, $CrPS_4$ characterised by the Néel temperature of 38 K. We combine Raman magnetospectroscopy with SQUID magnetometry, X-ray diffraction (XRD) and transmission electron microscopy. We support interpretation of the experimental data with the Density Functional Theory calculations taking as an input lattice constants obtained from temperature-dependent XRD measurements.

We conduct our study on bulk samples synthesised using the vapour transport method [1]. We observe that the phonon modes in the Raman spectra of $CrPS_4$ are strongly linearly polarised, which we attribute to a monoclinic structure of the material. Selected phonon lines traced as a function of temperature in the absence of magnetic field show an abrupt change of the spectral position, linewidth, and intensity upon crossing of the Néel temperature. The occurrence of a magnetic phase transition manifested in the Raman scattering spectrum is further corroborated by the maximum in magnetic susceptibility obtained from magnetometry measurements and the minimum of the unit cell volume determined by XRD.

Magnetic field induces a spectral shift of selected Raman lines, described by an approximately linear dependence. The slope of this dependence determined for consecutive temperatures in the range from 5 K to 150 K follows directly the magnetic susceptibility curve obtained from the magnetometry in this temperature range. In addition, the magnetic field increases the degree of circular polarization of the Raman lines. The impact of the field on the degree of circular polarisation is significant below the Néel temperature and becomes much weaker upon a transition to the paramagnetic phase.

Our work provides evidence that the Raman scattering, thus a purely spectroscopic technique, enables the determination of such magnetic-type property of $CrPS_4$ as the magnetic susceptibility curve. Our calculations of phonon shifts taking into account spin ordering of Cr cations indicate which transitions in the Raman spectrum are optimal for that purpose. The calculations clarify also energy order of possible spin arrangements and determine values of exchange constants between Cr ions in the $CrPS_4$ lattice.

In a view of our results, Raman spectroscopy appears to be a remarkably efficient method for gaining insight into the magnetic order and critical behaviour of magnetic semiconductors. The magnetospectroscopy approach to determination of magnetic properties are particularly useful in the case of a single- or a few-layer samples of antiferromagnetic layered materials, for which the Raman scattering signal is still sizeable, while low signal-to-noise ratio precludes any magnetometry or neutron scattering measurements.

References

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