Magnetic structure and interactions in 2D layered van der Waals semiconductors $CrPS_4$ and $MnPSe_3$ probed with neutron scattering

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Two-dimensional (2D) materials are of intense current fundamental and applied interest as a route to create novel fundamental phenomena beyond well-established classical behaviour within their topologically constrained layers. In this context 2D monolayer graphene, formed from the isolation of weakly connected van der Waals (vdW) bonded 2D layers in graphite by exfoliation, ignited widespread interest. Exotic quantum relativistic phenomena, such as Dirac semi-metals and quantum anomalous Hall insulators, have been predicted in graphene and related materials ranging from isolated 2D monolayers to quasi-2D bulk materials with vdW bonded layers. The focus has expanded to "beyond graphene" 2D vdW layered materials with intrinsic properties such as magnetism and semiconductivity not present in graphene, however the number of materials is limited and detailed understanding only just beginning.

$MnPSe_3$ and $CrPS_4$ are such layered vdW materials that are both magnetic and semiconducting, with magnetic ions forming hexagonal and rectangular 2D motifs. To access their low dimensional behaviour we probe bulk powder and single crystal samples with neutron scattering measurements [1,2]. Through magnetic symmetry analysis and spin wave analysis we are able to isolate and explore the 2D structural and magnetic behaviour in these bulk materials. Interactions shown in Fig. 1. The data highlights subtle competing interactions in both materials that leads to the stabilization of the determined magnetic ground states. These magnetic ground states were further tuned with small applied perturbations of field and temperature and found to undergo both subtle spin alterations and more dramatic metamagnetic transitions. The determination of the intralayer and interlayer exchange interactions and anisotropy within model spin Hamiltonians allowed the underlying observed exotic bulk behaviour to be explored.

The results show that for $MnPSe_3$ the Se ion drives unexpectedly strong magnetic interactions between the 2D layers, which forms a contrast to the wider studies S analogue $MnPS_3$. While for $CrPS_4$ a further lowering of interaction dimensionality to 1D-chains is shown to be of significance. Collectively, these results highlight the subtle role of the crystalline structure on the emergent behaviour and show the powerful insights neutron scattering can supply to studies of low dimensional materials.

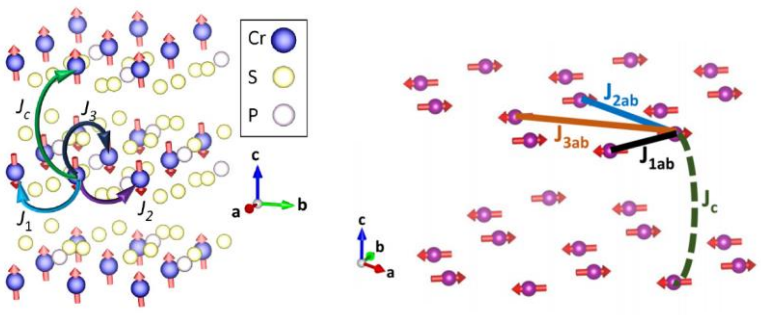

Figure 1. Magnetic structure and exchange interaction for the 2D layered materials $CrPS_4$ and $MnPSe_3$.

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