Magnetic properties of few-layer $Cr_2Ge_2Te_6$: A comparative ab initio study

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Atomically-thin magnetic crystals[1-4] have been recently isolated experimentally, greatly expanding the family of two-dimensional (2D) materials.

In this work we focus our attention on $Cr_2Ge_2Te_6$, presenting an extensive comparative analysis of its magnetic properties, based on ab initio density-functional theory (DFT)[5-7]. We first show that the often-used DFT+U fails in predicting the ground-state properties of this material in both its monolayer and bilayer forms, and even more spectacularly in its bulk form. On the contrary, the use of hybrid functionals, which naturally take into account correlations between all orbitals (and not only between the d orbitals of Cr), yields very good account of the available experimental data.

We then calculate all the relevant exchange couplings for mono- and bi-layer $Cr_2Ge_2Te_6$ and its bulk form, comparing results obtained via different methods and/or supercell size. In the case of bilayer Cr2Ge2Te6, we show that two distinct second-neighbor exchange couplings emerge.

We have tried to give a detailed account of all available results in the literature (including also those for the magneto-crystalline anisotropy), comparing them with our findings, and commenting on the discrepancies.

References

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