Evaluating critical temperatures for 2D ferromagnetic order: Beyond the random phase approximation

Varun Rajeev Pavizhakumari
Thorbjørn Skovhus, Thomas Olsen
CAMD, Computational Atomic-Scale Materials Design, Department of Physics, Technical University of Denmark, 2800 Kgs. Lyngby Denmark
vrapa@dtu.dk

Abstract

This research work investigates thermodynamic properties of magnetic materials using the Heisenberg model with localized interacting spins. As the mean field theory for magnetism fails in 2D by contradicting Mermin-Wagner theorem, we resort to the Green's function methods and the Holstein-Primakoff(HP) bosonization method. Even though the Green's function based Random Phase Approximation(RPA) is a common approach to determine critical temperatures, it underestimates the critical temperature for larger spin regimes and predicts the wrong ground state behavior for 2D materials. In this work, we try to compare the performance of RPA to a much more accurate scheme for Green's function method proposed by Herbert Callen[1] and as well as to HP bosonization with interacting magnons for ferromagnetic systems in both bulk and 2D.

We start by considering a model cubic lattice in the nearest neighbor limit and show that critical temperature from the Callen's scheme and HP scheme with higher order interacting terms in the classical limit has very close agreement to the classical Monte Carlo(MC) simulations. Then these methods are applied to the cases of bcc Fe, fcc Co and fcc Ni with exchange interactions calculated using first principle based Magnetic Force Theorem. Here too, we observe that Callen's method predicts closer agreement to the experimental critical temperatures whereas the conventional RPA underestimates the value every time.

Finally, we consider 2D materials where magnetic anisotropy is crucial for magnetic order. We first address the ambiguity of single ion anisotropy associated with RPA. The treatment of single ion anisotropy with Callen's method and HP bosons with higher order terms predicts the lack of order in S=1/2 materials whereas RPA fails. For nearest neighbour 2D lattices(square, hexagonal and honeycomb), we follow the same approach as in cubic by comparing the classical limit of critical temperature by all methods to the classical MC simulations for a range of anisotropies. To validate the predictions, we apply these methods to the monolayer CrI3. Considering the ambiguity in first principle exchange parameters of CrI3, we rely on experimental magnon energy and spin wave gap to obtain the exchange parameters. We observe that the HP method yields closer agreement to the experimental critical temperature of 45 K.

In conclusion, we have observed that the Callen's method exhibits a clear advantage over RPA for bulk metallic ferromagnetic materials and for 2D magnetic materials, HP bosonization and Callen's method can be potentially better candidates than RPA to predict critical temperatures.

References

[1] Herbert B Callen, Physical Review, 130 (1963) 890