Thermodynamics of magnetic systems from first principles: Combining Monte Carlo and Density Functional calculations

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Density functional calculations have proven to be a useful tool in the study of ground state properties of many materials. The investigation of finite temperature magnetism on the other hand has to rely usually on the usage of empirical models that allow the large number of evaluations of the system's Hamiltonian that are required to obtain the phase space sampling needed to obtain the free energy, specific heat, magnetization, susceptibility, and other quantities as function of temperature. We have demonstrated a solution to this problem that harnesses the computational power of today's large massively parallel computers by combining a classical Wang-Landau Monte-Carlo calculation [F. Wang and D. P. Landau, PRL 86, 2050 (2001)] with our first principles multiple scattering electronic structure code (LSMS) that allows the energy calculation of constrained magnetic states [M. Eisenbach et al., SC'09: Proceedings of the Conference on High Performance Computing, Networking, Storage and Analysis, ACM (2009)]. We present our calculations of finite temperature properties of Fe and Fe_3C using this approach and we find the Curie temperatures to be 980K and 400K respectively, in good agreement with experiments. [M. Eisenbach et al., J. Appl. Phys. 109, 07E138 (2011)] With current computational resources it is not yet feasible to combine both the atomic displacements and the magnetic order in a first principles method as described above. To investigate the interplay of these two subsystems we derive effective classical models based on our first principles calculations that allow us to treat the positional and magnetic degrees of freedom on and equal footing within classical Monte Carlo simulations. The models thus obtained give remarkably good agreement for thermodynamic and magnetic properties of iron between 300K and the Curie temperature. [J. Yin et al., PRB 86, 214423 (2012)]

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