Calculations of Magnetic States of Transition Metal Clusters Using a Variational Implementation of a Self-Interaction Corrected Energy Functional

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While Kohn-Sham density functional theory (KS-DFT) has been extremely successful and is now widely used in the studies of molecules and condensed matter it has several shortcomings. This is mainly due to the self-interaction error (SIE) inherent to practical implementations of the method. It, for example, often leads to an underestimation of the magnetic moment of metallic clusters. The Perdew-Zunger self-interaction correction (PZ-SIC) method is a way to correct for the spurious SIE [1]. Calculations based on PZ-SIC can describe challenging systems where conventional exchange-correlation functionals fail. These systems include defect states in crystals [2], charge-localized states in molecules [3] and diffuse states with a weakly bound electron [4-5]. In this study PZ-SIC is applied in calculations of magnetic states of small clusters of iron, cobalt and nickel, and is shown to improve the agreement with experiments compared to KS-DFT. Fe_{13} is anomalous as it appears to be antiferromagnetic while the other Fe clusters are ferromagnetic. The reason for this is not known but could be related to the presence of impurities that destabilize the symmetric icosahedral structure.

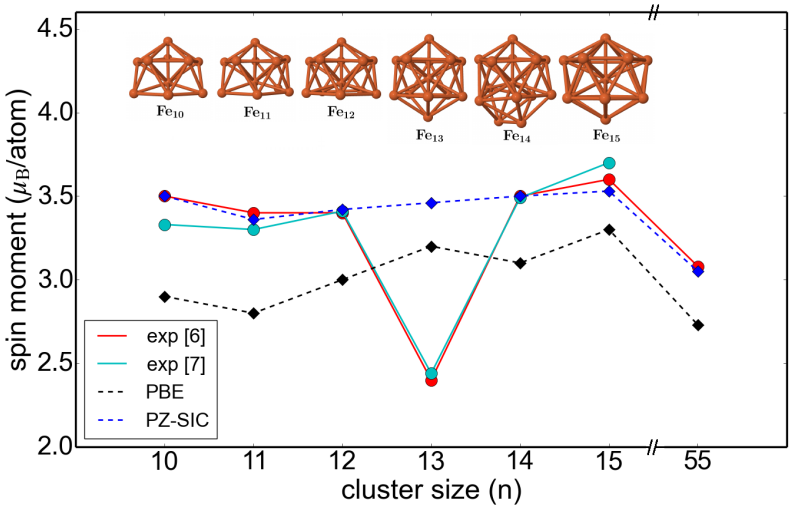

Figure 1: Experimental and theoretical magnetic moment per atom in Fe_n clusters as a function of size, calculated with PBE and PZ–SIC applied to PW91. PZ–SIC greatly improves upon the PBE results, and gives good agreement with the experimental results, except for Fe_{13} which is anomalous.

[1] J. P. Perdew and A. Zunger Phys. Rev. B 23, 5048 (1981)
[2] H. Guðmundsdóttir, E. Ö. Jónsson and H. Jónsson N. J. Phys. 17, 083006 (2015)
[3] X. Cheng, Y. Zhang, E. Ö. Jónsson, H. Jónsson and P. M. Weber Nat. Commun. 7, 11013 (2016)
[4] H. Guðmundsdóttir, Y. Zhang, P. M. Weber and H. Jónsson J. Chem. Phys. 141, 234308 (2014)
[5] Y. Zhang, P.M. Weber. and H. Jónsson J. Phys. Chem. Letters 7, 2068 (2016).
[6] M. Niemeyer *et. al.* Phys. Rev. Lett. 108, 057201 (2012).
[7] A. Langenberg *et. al.* Phys. Rev. B 90, 184420 (2014).