Magnetic Properties of Transition metal nanoparticles: A DFT-Inhomogeneous-DMFT analysis

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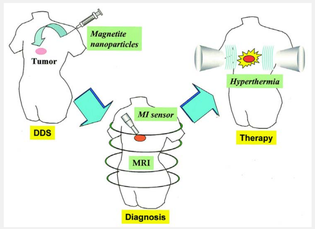
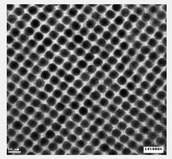
1 Motivation

Magnetic transition metal (TM) clusters are important ingredients in many technological applications from nanomedicine to recording media.



Akira Ito, et al. J. Biosci. Bioeng.: 100, 1(2005)

FePt SOMA media

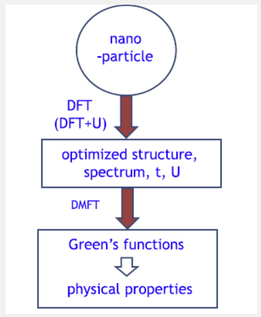


6.3+/-0.3 nm FePt particles

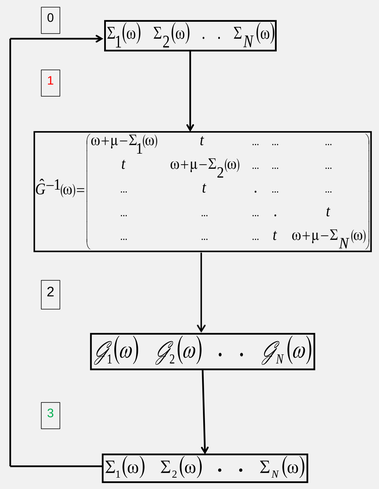
Shouheng Sun, et al. Science 287, 1989(200)

DFT+U approach overcomes some major deficiencies of DFT but it is still one-electron method because it is based on static mean filed approximation. It completely fails for strongly correlated metals. It cannot also describe paramagnetic insulator because it works only for long-range spin and orbital order.

3 nanoDMFT scheme



Turkowski, Kabir, Nayyar, Rahman, J. Phys. Cond. Mat. 22, 462202 (2010)



Despite zero dimensionality, DMFT approximation is valid in the case of nanostructures if the coordination number is large

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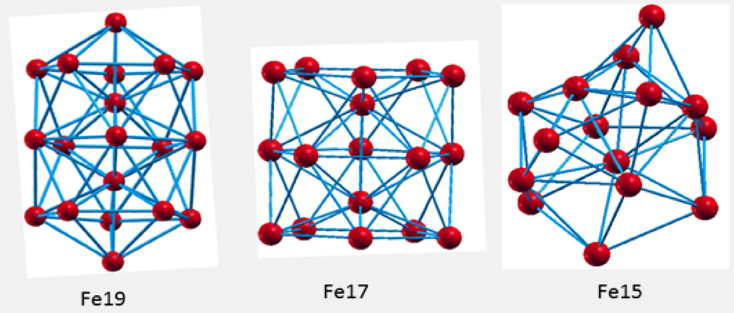
The structures optimized within the GGA+U approximation:



Quantum Espresso:J.Phys.:Condens.Matter,21, 395502 (2009).

For the effective Hubbard Hamiltonian, the hopping parameters were used in the form of Slater-Koster inter-atomic matrix elements with following s- and d-orbital couplings:

$$V_{ss\sigma} = -\frac{\pi^2}{8}\frac{\hbar^2}{md^2},\quad V_{sd\sigma} = -\frac{1}{\pi}\sqrt{\frac{5}{4}}\frac{\hbar^2 r_d^{3/2}}{md^{7/2}},$$

$$V_{dd\sigma} = -\frac{45}{\pi}\frac{\hbar^2 r_d^3}{md^5},\quad V_{dd\pi} = \frac{30}{\pi}\frac{\hbar^2 r_d^3}{md^5},\quad V_{dd\delta} = -\frac{15}{2\pi}\frac{\hbar^2 r_d^3}{md^5},$$

where r_d is the average d-state radius and d is the inter-atomic distance.

2 DFT approach

Choose initial electronic density $\rho(r)$

Calculate effective Potential using the LDA equation
$V_{eff}(r) = V_{ion}(r) + \int d^3r' V_{ee}(r-r')\rho(r') + \frac{\delta E_{xc}[\rho]}{\delta\rho(r)}$

Solve Kohn-Sham equation
$\left[-\frac{\hbar^2}{2m}\Delta + V_{eff}(r) - \varepsilon_i\right]\varphi_i(r) = 0$

Calculate electronic density
$\rho(r) = \sum_i^N |\varphi_i(r)|^2$

DFT+DMFT scheme

Choose an initial self energy Σ

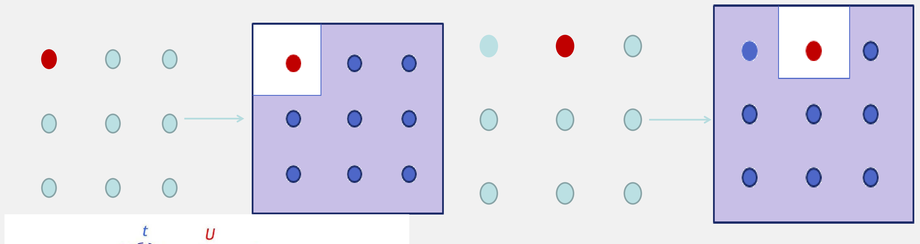
Calculate Green function from Σ
$G_{qlm,q'l'm'}(\omega) = \frac{1}{V_B}\int d^3k\left(\left[\omega 1 + \mu 1 - H_{GGA}(K) - \Sigma(\omega)\right]^{-1}\right)_{qlm,q'l'm'}$

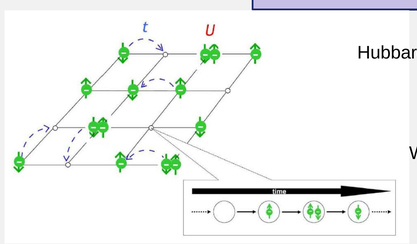
$g = (G^{-1} + \Sigma)^{-1}$

Calculate G via the DMFT single-site problem
$G^{\sigma}_{\nu m} = -\frac{1}{Z}\int D[\psi]D[\psi^*]\psi^{\sigma}_{\nu m}\psi^{\sigma *}_{\nu m} e^{A[\psi,\psi^*,g^{-1}]}$

Calculate new self energy
$\Sigma_{new} = g^{-1} - G^{-1}$

G. Kotliar et al, Rev. Mod. Phys. 78, 865 (2006)





Hubbard Model is the basic model to study the correlation effects

$$H = \sum_{i,j,\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U\sum_i n_{i\uparrow} n_{i\downarrow}$$

We applied Greens function technique to solve this Hamiltonian

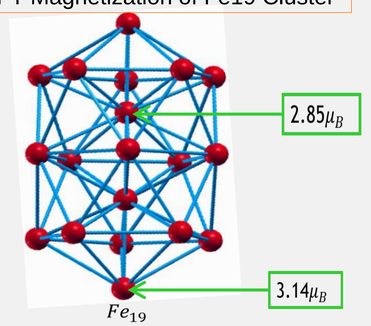
$$G(k,\omega) = \frac{1}{i\omega - k^2 + \mu - \Sigma(k,\omega)}$$

The magnetization on site i can be obtained from

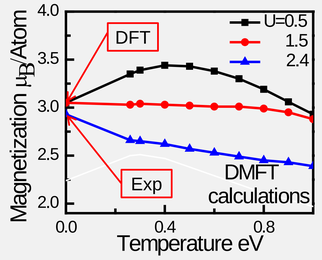
$$M_i(t) = -\mathrm{Im}\sum_{lm}\left[G_{ilm\uparrow,ilm\uparrow}(t,t+\delta^+) - G_{ilm\downarrow,ilm\downarrow}(t,t+\delta^+)\right]$$

5 Results

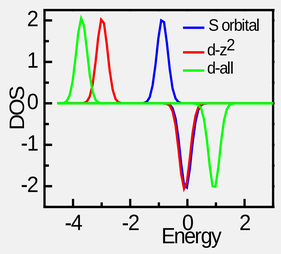
DFT Magnetization of Fe19 Cluster



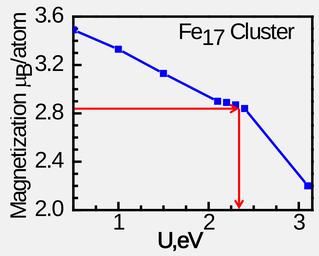
DMFT Magnetization of Fe19 Cluster



DOS of single Fe atom



DMFT Magnetization of Fe17 Cluster



magnetization obtained with different approaches

Clusters	DFT	DMFT	DFT+U	Experimental
Fe15	3.20	2.76(U=2.2)	3.15(U=2.2)	2.72
Fe17	3.06	2.87(U=2.3)	3.09(U=2.3)	2.86
Fe19	3.05	2.94(U=2.4)	3.07(U=2.4)	2.92

In most cases, DFT+U gives even more overestimated values of the magnetization than DFT. Dynamical fluctuation effects taken into account in DMFT lead to decrease of the magnetization, and the experimental results can be reproduced.

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Conclusions

- We show that similar to the case of bulk and 2D systems, this approach is a promising method to take into account the correlation effects even in the case of small clusters.
- It follows from our calculations that dynamical correlation effects lead to a significant decrease of the magnetization than DFT+U.
- DFT+DMFT gives better agreement with experimental values of magnetic moment than DFT+U.

Future Works

Generalization of the GGA+DMFT approach to the case of bimetallic cluster on substrates and to clusters with more atoms will be performed in the near future.

Acknowledgements:

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