Quantum Mechanical Simulation of Nano-composite Magnets on CRAY XT3

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Magnetic Nanoparticle Composites

Nanoscale magnetic crystallites formed in an amorphous or crystalline matrix or on a surface

- **Properties:**
  - Atomic scale effects play dominant role.
  - Physical parameters can be adjusted selectively.

- **The advantages:** an increased sensitivity to detect changes of magnetic field and a higher working temperature range.

- **Potential applications:** sensors, data storage, MRI contrast enhancing reagents, smart drug delivery, etc.
Science of Disk Drives

110 Gbit/in²

Inductive Write Element

GMR Read Sensor

B = 30 nm (σ_j < 3 nm)
W = 194 nm, t = 15 nm, d = 10 nm

Grain Structure and Magnetic Transition

~80-100 grains per bit

Direction of Disk Motion

Track of Recording Media

W = 194 nm, t = 15 nm, d = 10 nm
Present disk drives (~ 100 Gb/in²)
Use a metallic thin-film medium whose magnetic grains (~ 100 grains/bit), acting like an array of permanent magnet particles, are partially isolated from one another by a nonmagnetic chromium-rich alloy.

Superparamagnetic Limit (~ 100-200 Gb/in²)
A factor of 2 decrease in grain diameter ➔ a factor of 8 decrease of grain volume ➔ a factor of 8 decrease of magnetic energy stored in each grain ➔ a change of the magnetic moment reversal time, due to thermal fluctuation, from 100 years to as much as 100 nanoseconds.

Future magnetic data storage (~ 1Tb/in²)
- High magnetic anisotropy multilayers (Co/Pd, or FePt film in L1₀ phase)
- Perpendicular recording
- Magnetic nanoparticle composites with high magnetic anisotropy and 1 particle/bit:
  FePt, CoPt, etc.
Theoretical Approach

- **Empirical**
  - Model potentials for inter-atomic interactions
  - Less accuracy and poor predictability

- **Semi-empirical**
  - Parameters in the model potential are determined quantum mechanically
  - Better accuracy and reasonable predictability

- **Quantum mechanical (ab initio)**
  - No models
  - The best accuracy and predictability
Quantum Mechanical Solution of Materials Science Problems

Many-electron problem

Electron:  
Nucleus:  

Electron-electron interaction
Electron-nucleus interaction
Many-electron Schrödinger equation

One-electron problem

Density Functional Theory

Non-interacting electrons move in an effective potential: \( V_{\text{eff}}[\rho] \)

One-electron Schrödinger equation

\[
\left\{-\frac{\hbar^2}{2m_e} \nabla^2 + e^2 \int_\infty \frac{\rho(r')}{|\vec{r} - \vec{r}'|} d^3 r' - e^2 \sum_{\vec{R}_n} \frac{Z_n}{|\vec{r} - \vec{R}_n|} + V_{\text{xc}}[\rho]\right\} \Psi_\alpha(\vec{r}) = \varepsilon_\alpha \Psi_\alpha(\vec{r})
\]
Self-consistent Process

\[ V_{\text{eff}}(\vec{r}), \text{lattice parameters} \]

\[ \text{One-electron Schrödinger Equation} \]

\[ [-\nabla^2 + V_{\text{eff}}(\vec{r})] \cdot \Psi_{\alpha}(\vec{r}) = \epsilon_{\alpha} \Psi_{\alpha}(\vec{r}) \]

\[ E[\rho], \text{ etc.} \]

\[ \rho(\vec{r}) = \sum_{\alpha} |\Psi_{\alpha}(\vec{r})|^2 \]

\[ \text{LDA Potential} \]

\[ V_{\text{eff}}(\vec{r}) = e^2 \int_{\infty} \frac{\rho(\vec{r}')}{|\vec{r} - \vec{r}'|} d^3 r' - e^2 \sum_{R_n} \frac{Z_n}{|\vec{r} - \vec{R}_n|} + V_{\text{XC}}^{\text{LDA}}[\rho] \]

\[ \rho(\vec{r}), \text{lattice parameters.} \]

Local Density Approximation (LDA) makes calculation of \( V_{\text{XC}}[\rho] \) feasible.
Computational Challenges

- A spherical particle of 3 nm in diameter ~ 2000 atoms
- Bottlenecks of conventional \textit{ab initio} electronic structure methods
  - $N^3$ scaling in computational requirement
  - $N^2$ scaling in memory requirement
  - Dominated by global operations
  - Lack of efficient parallel implementation scheme
  - Simulation size is limited to 100s atoms
Locally Self-consistent Multiple Scattering (LSMS) Method

- Real space multiple scattering approach

Electron Density
\[
\rho_i(\vec{r}) = -\frac{1}{\pi} \text{Im} \text{Tr} \int_{-\infty}^{\varepsilon_F} dz \cdot G_i(\vec{r}, \vec{r}; z)
\]

Moment Density
\[
\vec{m}_i(\vec{r}) = -\frac{1}{\pi} \text{Im} \text{Tr} \int_{-\infty}^{\varepsilon_F} dz \cdot \left[ G_i(\vec{r}, \vec{r}; z) \cdot \vec{\sigma} \right]
\]

Moment Orientation
\[
\vec{e}_i = \frac{\int_{\Omega_i} d^3 \vec{r} \cdot \vec{m}_i(\vec{r})}{\left| \int_{\Omega_i} d^3 \vec{r} \cdot \vec{m}_i(\vec{r}) \right|}
\]

- Solve the multiple scattering equations associated with atom \(i\) and compute the Green’s function \(G_i\), which depends on the single scattering \(t\)-matrix and the location of the atoms in the local region.

- Order-\(N\) scaling in time and space complexity
To obtain the Green’s function $G_i$ for atom $i$, one needs to compute:

$$\tau^{ii}(\varepsilon) = \begin{bmatrix}
    t_i^{-1}(\varepsilon) & g_{ij}(\varepsilon) & g_{ik}(\varepsilon) & g_{in}(\varepsilon) & g_{im}(\varepsilon) \\
    g_{ji}(\varepsilon) & t_j^{-1}(\varepsilon) & g_{jk}(\varepsilon) & g_{jn}(\varepsilon) & g_{jm}(\varepsilon) \\
    g_{ki}(\varepsilon) & g_{kj}(\varepsilon) & t_k^{-1}(\varepsilon) & g_{kn}(\varepsilon) & g_{km}(\varepsilon) \\
    g_{ni}(\varepsilon) & g_{nj}(\varepsilon) & g_{nk}(\varepsilon) & t_n^{-1}(\varepsilon) & g_{nm}(\varepsilon) \\
    g_{mi}(\varepsilon) & g_{mj}(\varepsilon) & g_{mk}(\varepsilon) & g_{mn}(\varepsilon) & t_m^{-1}(\varepsilon)
\end{bmatrix}^{-1}$$

LIZ Size $\times (l_{\text{max}} + 1)^2$
Algorithm and Communication Pattern

**N-atom Unit Cell**

![Diagram of N-atom Unit Cell]

**Local Interaction Zone (LIZ)**

![Diagram of Local Interaction Zone (LIZ)]

**Algorithm and Communication Pattern**

Input: \( \nu_i(\vec{r}) \)

Compute: \( t \)-matrix

Receive: \( t_j, t_k, t_m, t_n \)

Send: \( t_i \)

Result: \( \rho_i(\vec{r}) \)
Parallel Implementation

- **Intrinsic parallelism:**
  - the atoms in the unit cell
  - the energy points along the complex energy contour

- Atoms are distributed evenly among CPUs, and more importantly, unlike the previous version, the code allows multiple atoms mapped onto each CPU

- One-sided communications are used for getting the $t$-matrix from those neighboring atoms that are mapped onto other CPUs

- Parallel I/O of data in XDR, HDF, or machine dependent binary format
LSMS Performance on Cray-XT3 (bigben) at PSC

- Perfect Scaling (GFLOPS)
- Estimated Performance (GFLOPS)

- GFLOPS for the Run

8.09 TFLOPS on 2068 nodes
8.03 TFLOPS on 2048 nodes

Number of Atoms (Nodes)

Total Performance (GFLOPS)
Fe nanoparticle embedded in FeAl matrix
(16,000 Fe and Al atoms per unit cell)

Elapsed Time (sec)
Inverse law (sec)

Number of CPUs

- 8 atoms/CPU
- 10 atoms/CPU
- 16 atoms/CPU
- 20 atoms/CPU
- 25 atoms/CPU

Spin-polarized calculation
5 self-consistent iterations
LIZ = 89
Lmax = 3
Applications of LSMS Method

Electronic Structure of Magnetic Nanoparticles

- FePt nanoparticle (L1₀ structure) embedded in FePt (face-centered tetragonal structure) random alloy matrix
- No lattice mismatch ($a_0 = 3.8525\text{Å}$ and $c_0 = 3.7133\text{Å}$)
- Spin-polarized LSMS calculations are applied to the unit cell sample simulating the system
- The FePt nanoparticles of different sizes: 2.5nm, 3.86nm, and 5.0nm, each of which contains 711, 2,195, and 4,777 Fe and Pt atoms, respectively
- All together, there are 14,400 Fe and Pt atoms in the unit cell sample
Fe or Pt

Ordered compound in L1₀ structure

Pt

Fe

Random alloy in fct structure

Fe or Pt
Ferromagnetic Iron-Platinum (L1₀ phase) nanoparticle: 2,195 atoms, 3.86 nm in diameter

By Greg Foss at PSC
Ferromagnetic FePt nanoparticle (3.86 nm) embedded in FePt fct random alloy

Total simulation size: 14,400 atoms

By Greg Foss at PSC
FePt nanoparticle embedded in FePt random alloy

FePt (L1₀) nanoparticle (diameter = 38.6 Å)

FePt (fct) random alloy

excess electrons vs. distance from the center (Angstrom)
FePt nanoparticle embedded in FePt random alloy

magnetic moment (Bohr magneton)

FePt (L1₀)
nanoparticle
(diameter = 38.6)

FePt (fct) random alloy

distance from the center (Angstrom)
FePt nanoparticle embedded in FePt random alloy

Excess electrons on Fe and Pt as a function of the number of unlike nearest neighbors.
FePt nanoparticle embedded in FePt random alloy

- Magnetic moment on Fe
- Magnetic moment on Pt

![Graph showing magnetic moment vs number of unlike nearest neighbors for Fe and Pt]
FePt nanoparticle embedded in FePt random alloy

The graph shows the long range electrostatic potential (in atomic units) on the x-axis and the excess electrons on the y-axis. The graph compares the potential for Fe and Pt.

- The Fe line is represented by red dots and is positioned above the Pt line, indicating a higher potential for Fe.
- The Pt line is represented by blue squares and is positioned below the Fe line, indicating a lower potential for Pt.

The data points for Fe and Pt are spread across the graph, with Fe having a higher potential at the lower end of the x-axis and Pt having a lower potential at the upper end of the x-axis.
Conclusions

- The central region of the nanoparticle resembles the bulk properties.
- Dramatic changes from the bulk properties are seen in the surface region ~ 4 Å in width.
- The size effect on the charge and moment is small.
- Strong correlations between the number of unlike nearest neighbors and the electronic and magnetic properties.
- With the linear-scaling *ab initio* method and a 20 teraflop machine, we should be able to perform electronic structure calculations for nanoparticles of diameter size up to ~ 10nm (~ 80,000 atoms).
- It requires a petaflop machine to perform realistic simulations for nanoparticles of ~ 50nm (~ 5,000,000 atoms) in size, and other nanomaterials such as nanowire and nanotubes.
Questions to be Answered

- Interaction between the nanoparticles versus the distance, shape, etc
- Noncollinear magnetic structure at the interface region between the nanoparticles or between the nanoparticle and the surrounding matrix
- Size and shape effects on Magneto-anisotropy of the nanoparticle
- Relaxation in the interface region
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