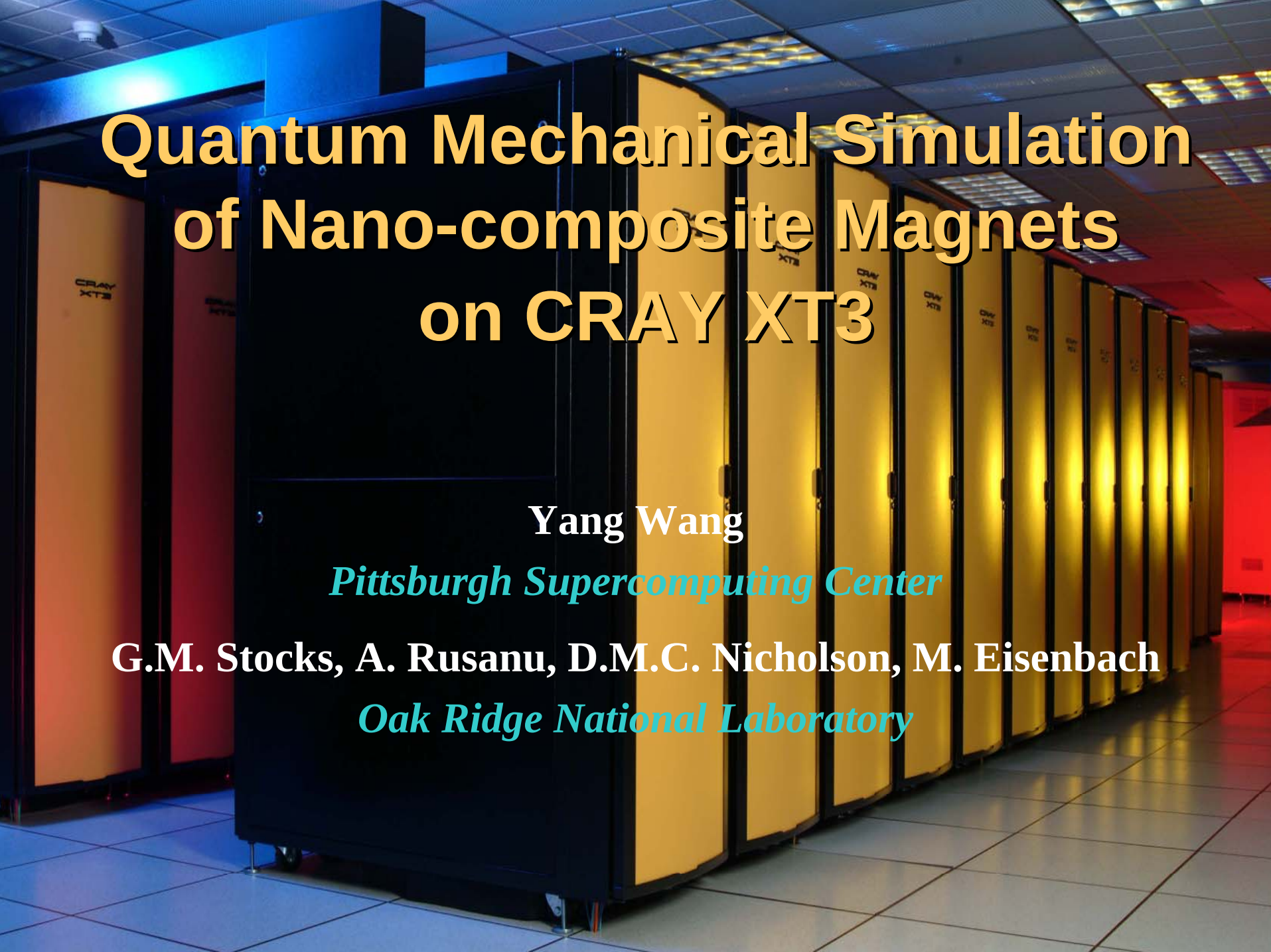

Quantum Mechanical Simulation of Nano-composite Magnets on CRAY XT3

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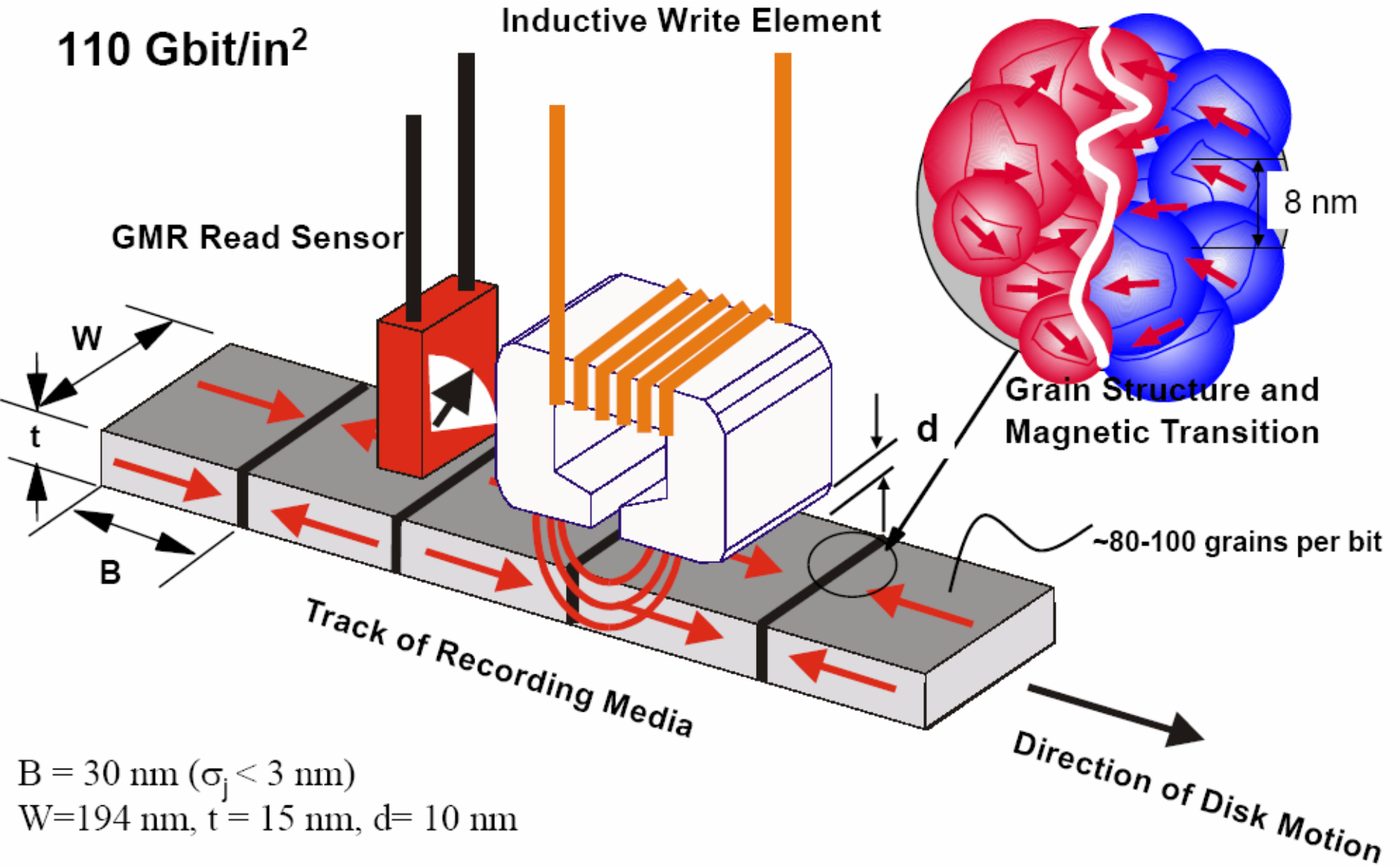
Magnetic Nanoparticle Composites

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



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

Pushing the Limit

- Present disk drives (~ 100 Gb/in^2)

  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/in^2)

  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/in^2)
  - High magnetic anisotropy multilayers (Co/Pd, or FePt film in $L1_0$ phase)
  - Perpendicular recording
  - Magnetic nanoparticle composites with high magnetic anisotropy and 1 particle/bit:

    FePt, CoPt, etc.

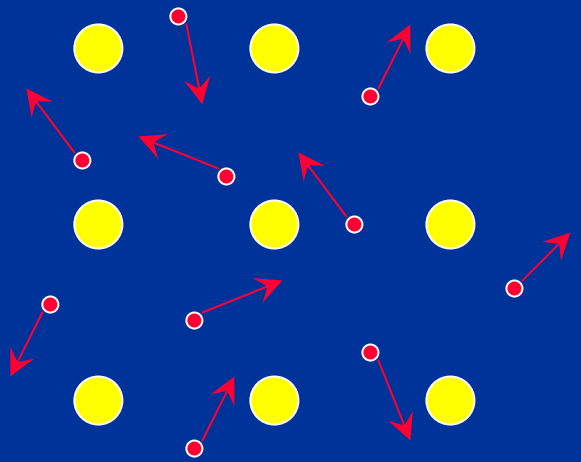
Theoretical Approach

- 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

Electron: **Nucleus:**

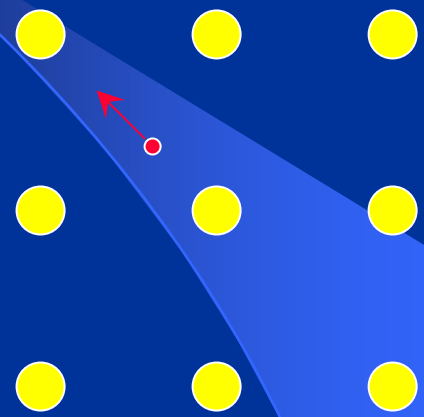
Many-electron problem



electron-electron interaction
electron-nucleus interaction
many-electron Schrödinger equation

Density Functional Theory

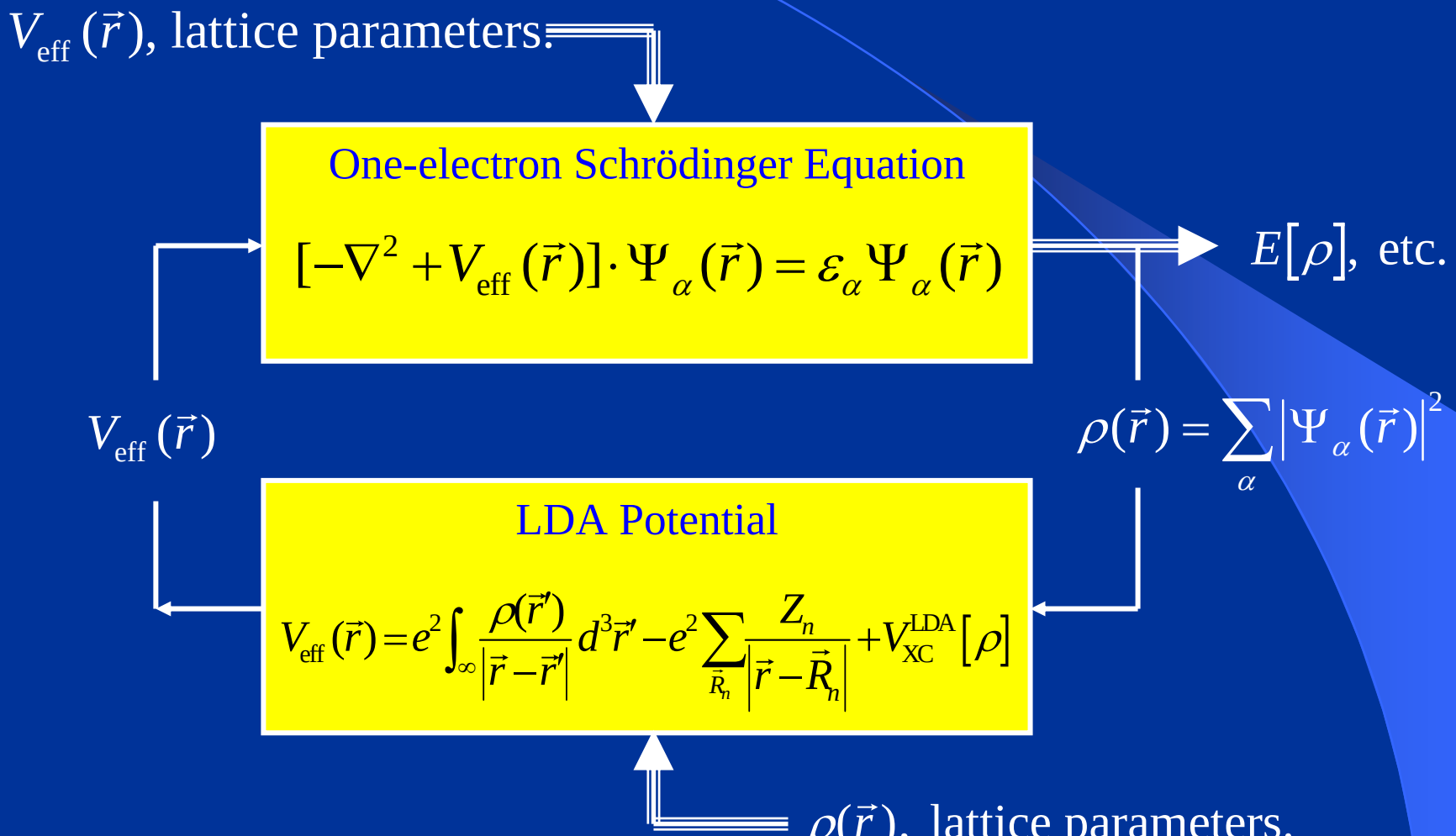
One-electron problem



non-interacting electrons move in an effective potential: $V_{\text{eff}}[\rho]$
one-electron Schrödinger equation

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

Self-consistent Process



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 *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 → $$\rho_i(\vec{r}) = -\frac{1}{\pi}\,\mathrm{Im}\,Tr\int_{-\infty}^{\varepsilon_F} dz\cdot \underline{G}_i(\vec{r},\vec{r};z)$$

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

Moment Orientation → $$\vec{e}_i = \int_{\Omega_i} d^3\vec{r}\cdot\vec{m}_i(\vec{r}) \Big/ \left|\int_{\Omega_i} d^3\vec{r}\cdot\vec{m}_i(\vec{r})\right|$$

- Solve the multiple scattering equations associated with atom ***i*** and compute the Green's function $\underline{\boldsymbol{G}}_{\boldsymbol{i}}$, 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

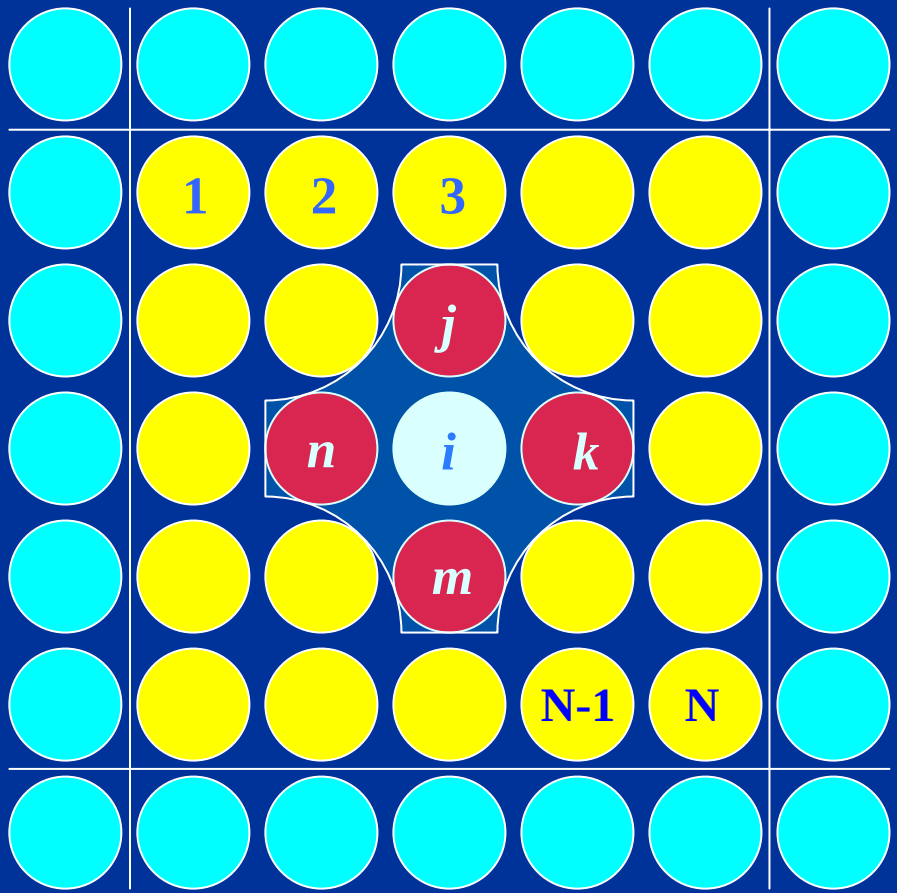
Major Floating-Point Operations

To obtain the Green's function $\underline{G}_i$ for atom i, one needs to compute:

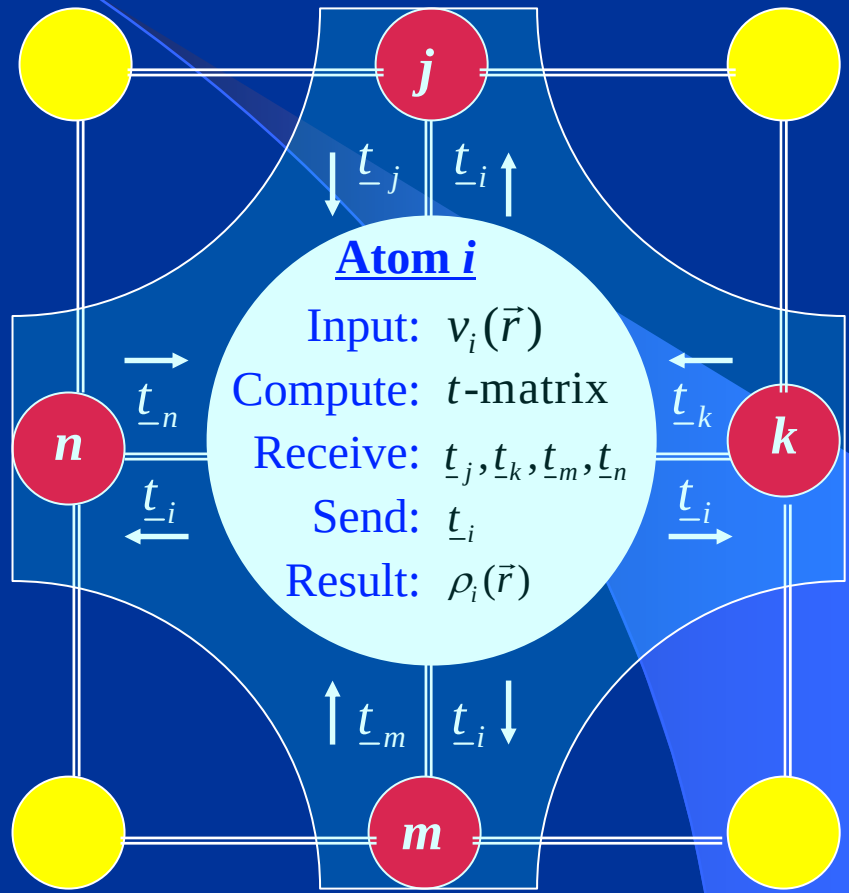
$$\underline{\tau}^{ii}(\varepsilon) = \underbrace{\begin{bmatrix} \underline{t}_i^{-1}(\varepsilon) & \underline{g}_{ij}(\varepsilon) & \underline{g}_{ik}(\varepsilon) & \underline{g}_{in}(\varepsilon) & \underline{g}_{im}(\varepsilon) \\ \underline{g}_{ji}(\varepsilon) & \underline{t}_j^{-1}(\varepsilon) & \underline{g}_{jk}(\varepsilon) & \underline{g}_{jn}(\varepsilon) & \underline{g}_{jm}(\varepsilon) \\ \underline{g}_{ki}(\varepsilon) & \underline{g}_{kj}(\varepsilon) & \underline{t}_k^{-1}(\varepsilon) & \underline{g}_{kn}(\varepsilon) & \underline{g}_{km}(\varepsilon) \\ \underline{g}_{ni}(\varepsilon) & \underline{g}_{nj}(\varepsilon) & \underline{g}_{nk}(\varepsilon) & \underline{t}_n^{-1}(\varepsilon) & \underline{g}_{nm}(\varepsilon) \\ \underline{g}_{mi}(\varepsilon) & \underline{g}_{mj}(\varepsilon) & \underline{g}_{mk}(\varepsilon) & \underline{g}_{mn}(\varepsilon) & \underline{t}_m^{-1}(\varepsilon) \end{bmatrix}^{-1}}_{\text{LIZ Size} \times (l_{\max}+1)^2}$$

11

Algorithm and Communication Pattern



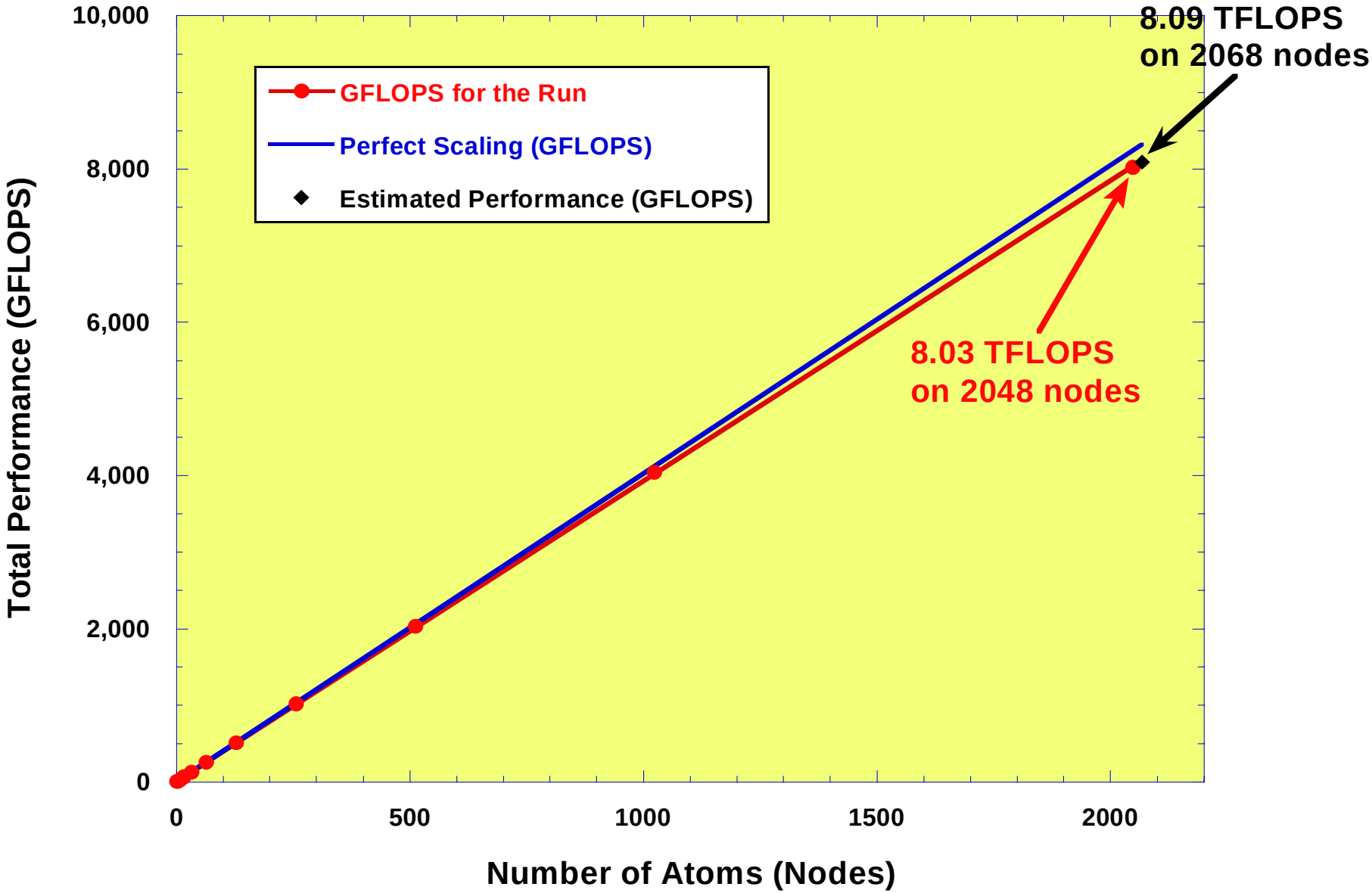
N-atom Unit Cell

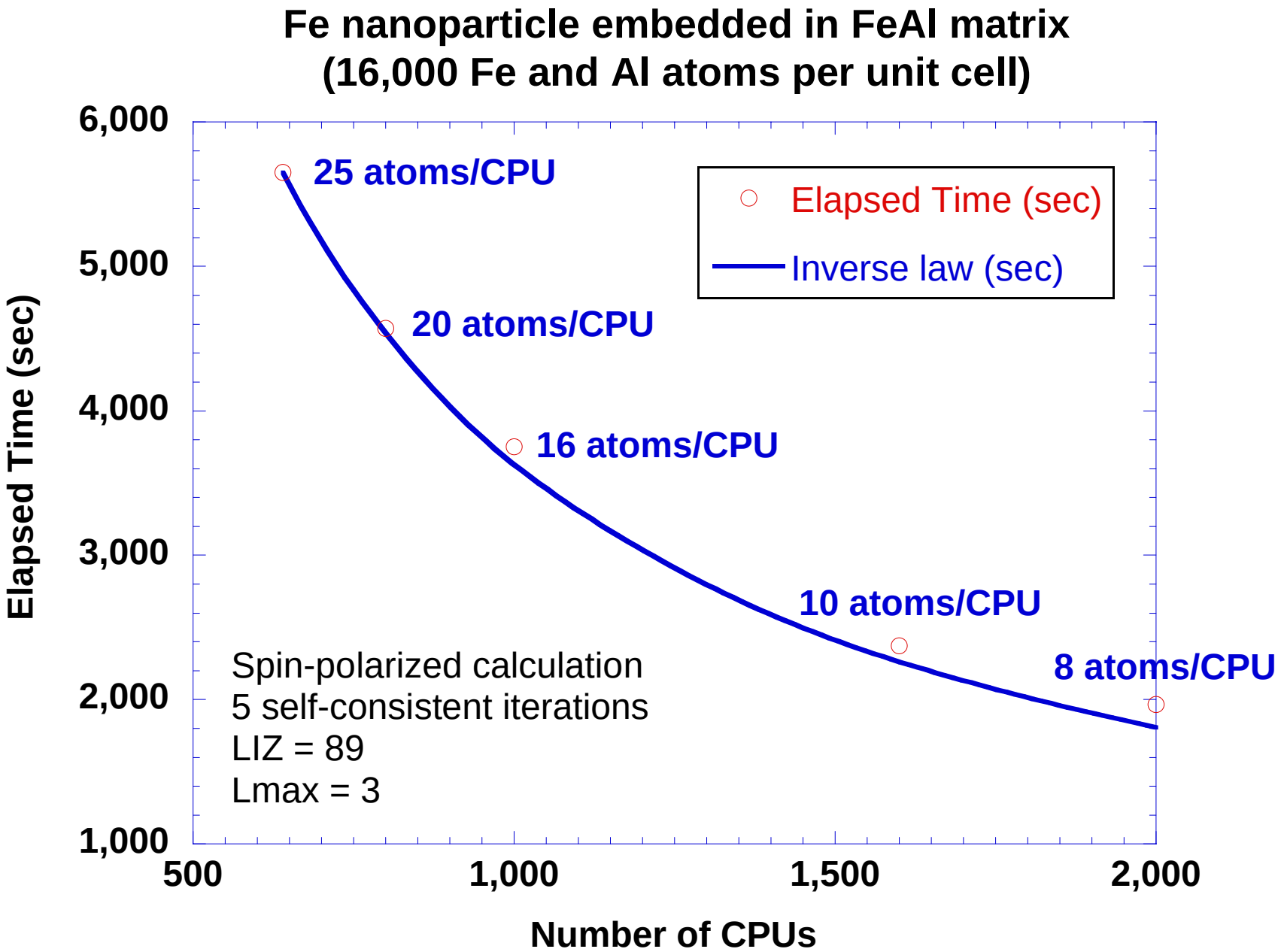


Local Interaction Zone (LIZ)

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
Total Performance (GFLOPS)
Number of Atoms (Nodes)
10,000
8,000
6,000
4,000
2,000
0
0
500
1000
1500
2000
GFLOPS for the Run
Perfect Scaling (GFLOPS)
Estimated Performance (GFLOPS)
8.09 TFLOPS
on 2068 nodes
8.03 TFLOPS
on 2048 nodes

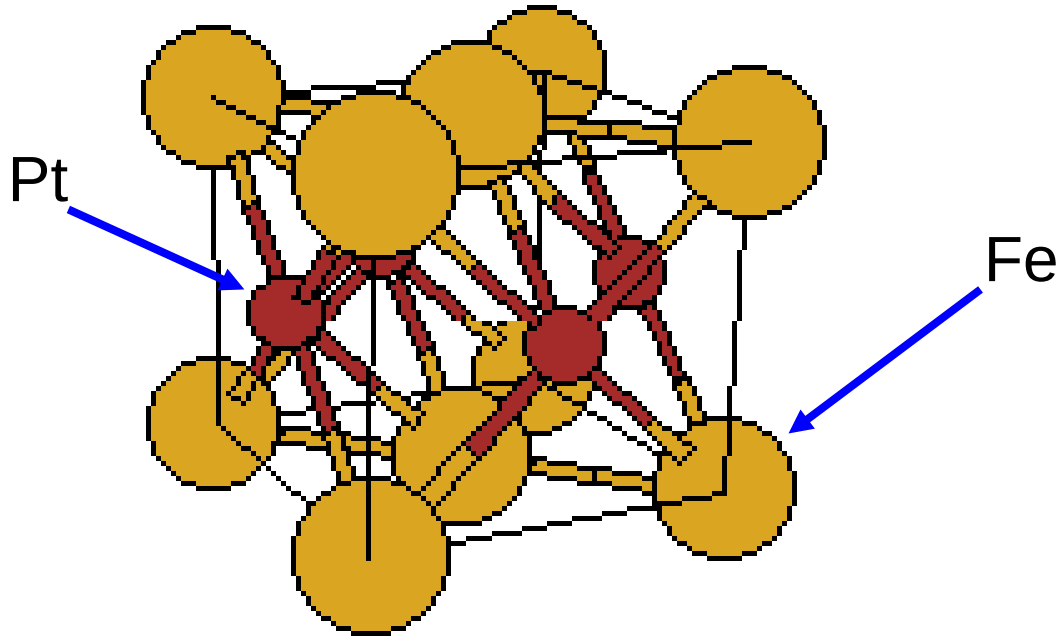

Fe nanoparticle embedded in FeAl matrix
(16,000 Fe and Al atoms per unit cell)
Elapsed Time (sec)
6,000
5,000
4,000
3,000
2,000
1,000
500
1,000
1,500
2,000
Number of CPUs
25 atoms/CPU
20 atoms/CPU
16 atoms/CPU
10 atoms/CPU
8 atoms/CPU
Elapsed Time (sec)
Inverse law (sec)
Spin-polarized calculation
5 self-consistent iterations
LIZ = 89
Lmax = 3

Applications of LSMS Method

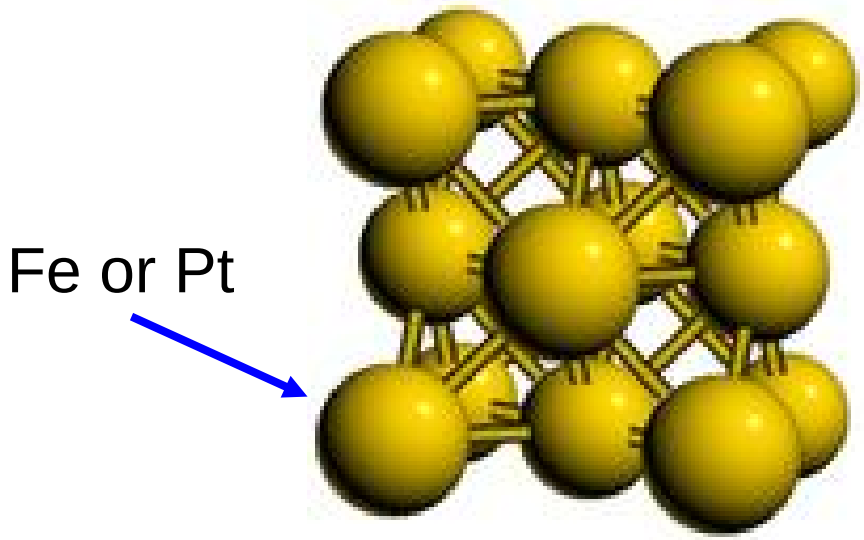
Electronic Structure of Magnetic Nanoparticles

- FePt nanoparticle ($L1_0$ structure) embedded in FePt (face-centered tetragonal structure) random alloy matrix
- No lattice mismatch (a_0 = 3.8525Å and c_0 = 3.7133Å)
- 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

Ordered compound in $L1_0$ structure



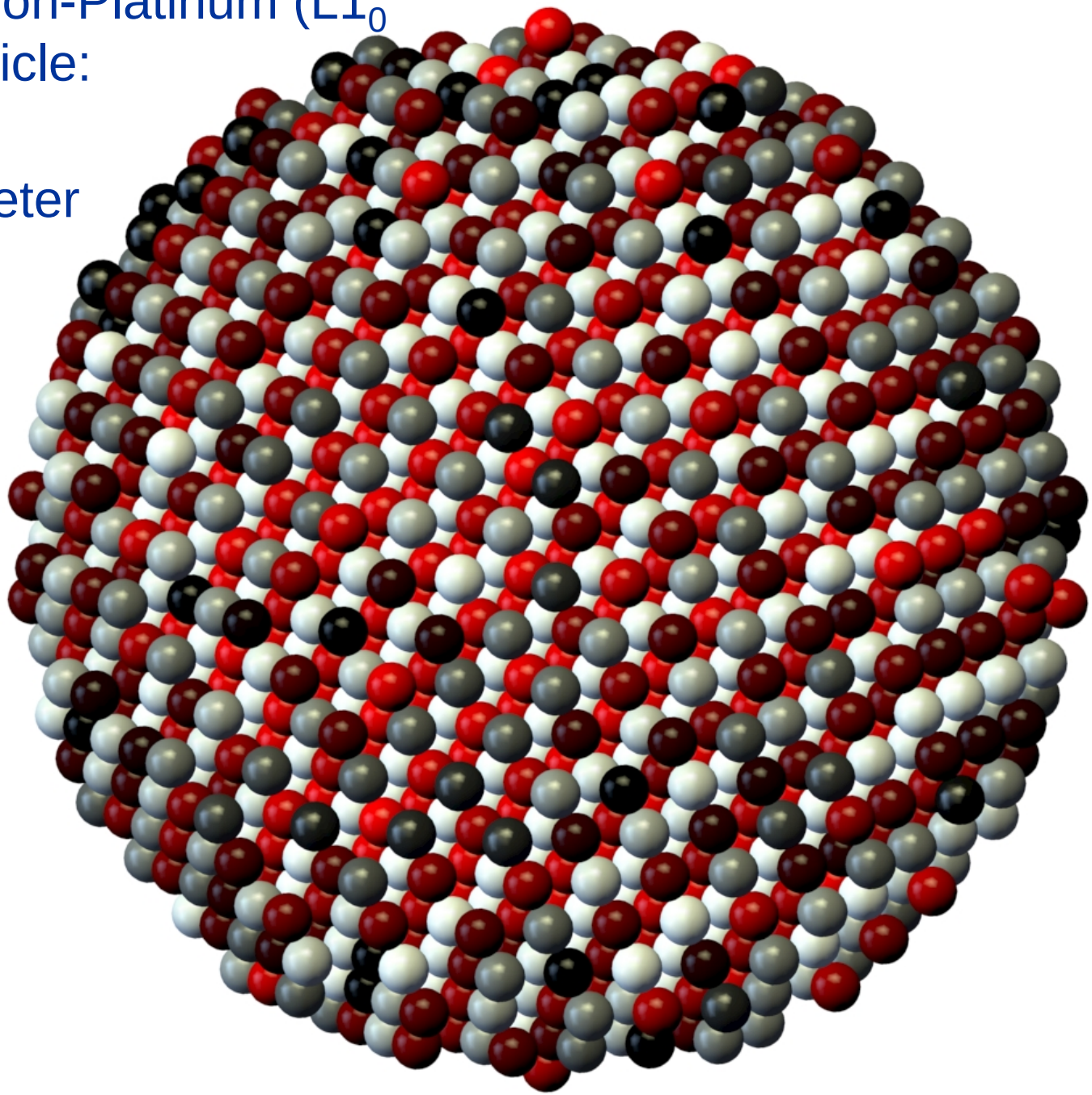
Random alloy in fct structure



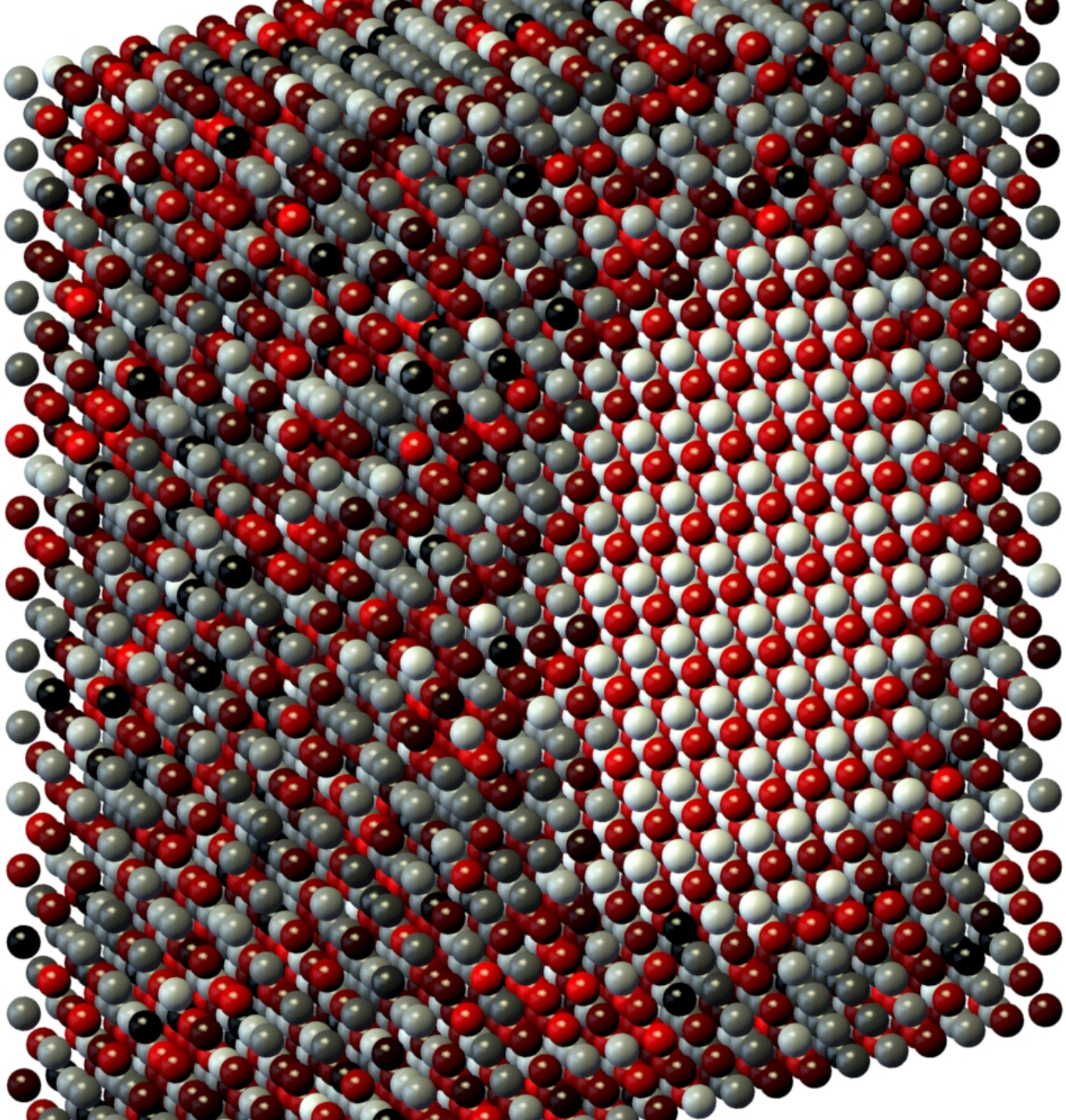
Ferromagnetic Iron-Platinum ($L1_0$ phase) nanoparticle:

2,195 atoms,

3.86 nm in diameter



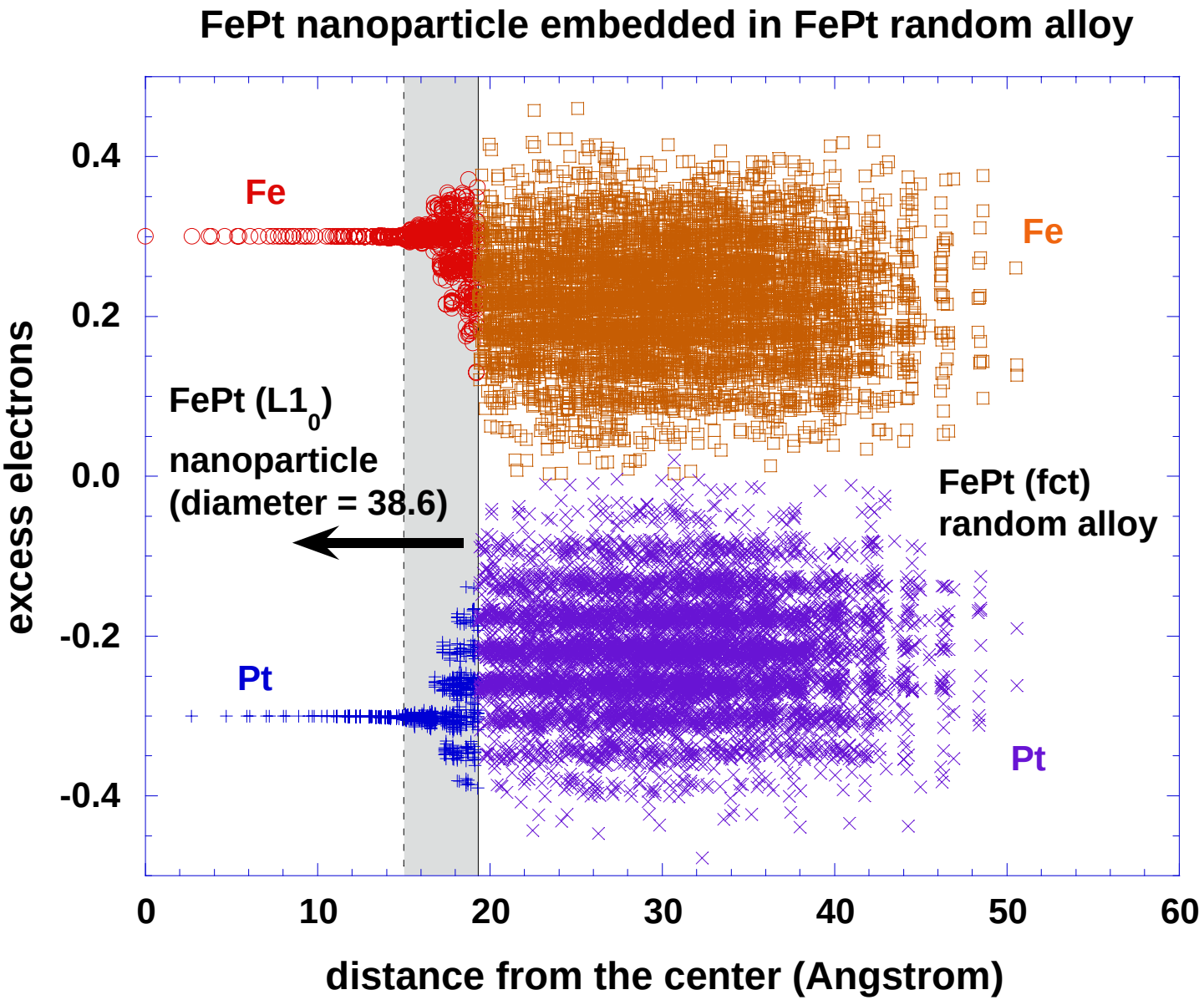
By Greg Foss at PSC

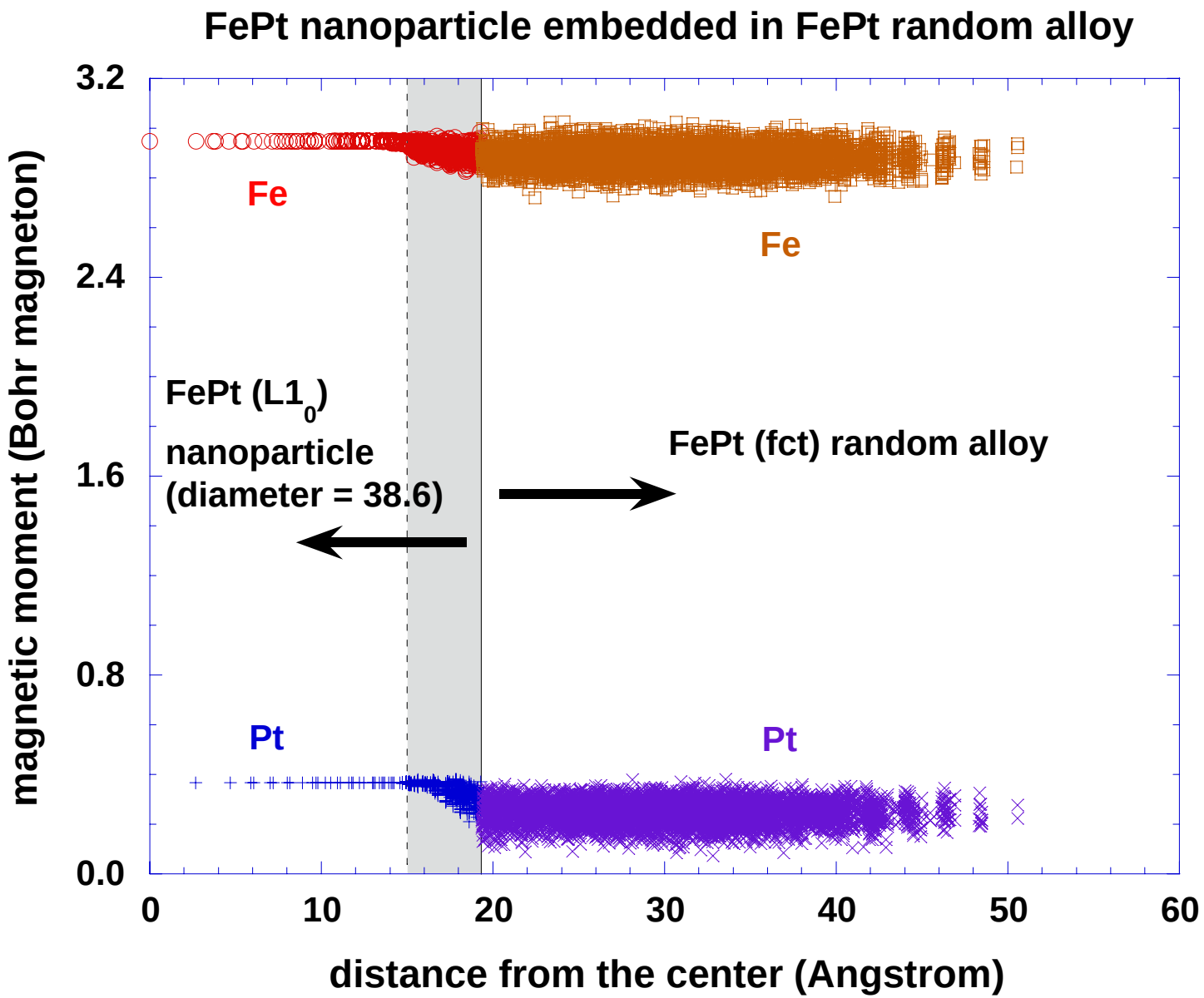


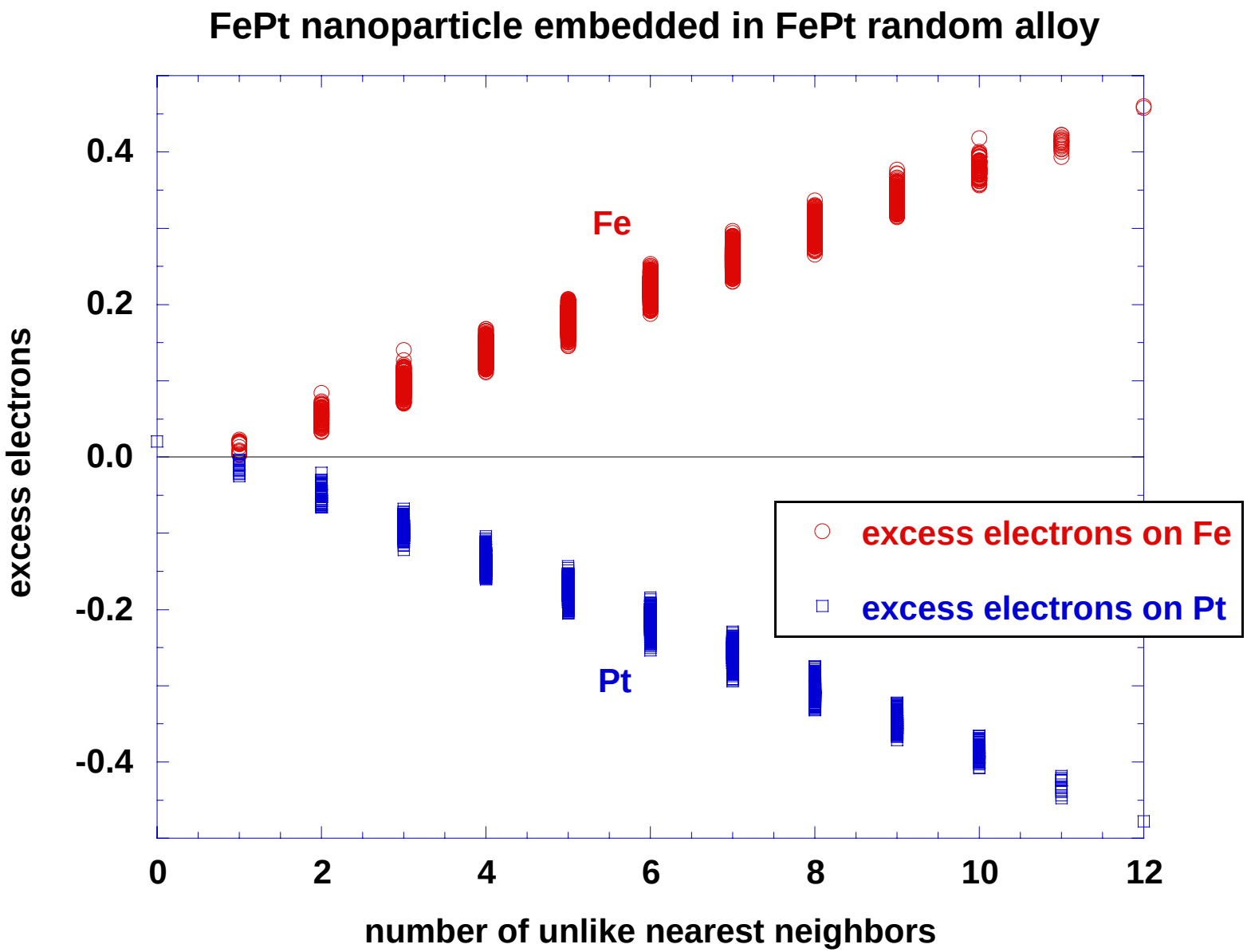
Ferromagnetic FePt nanoparticle (3.86 nm) embedded in FePt fct random alloy

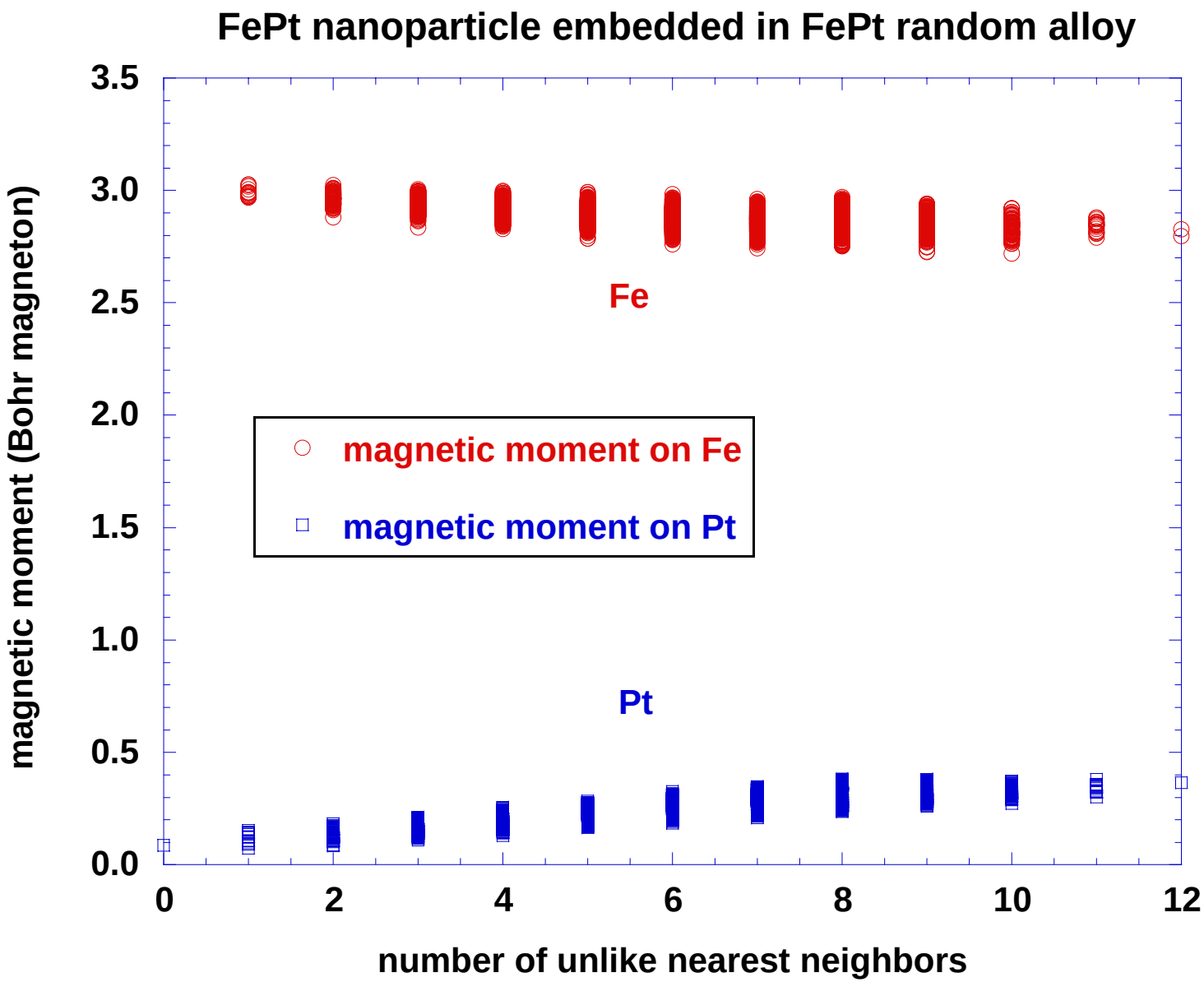
Total simulation size: 14,400 atoms

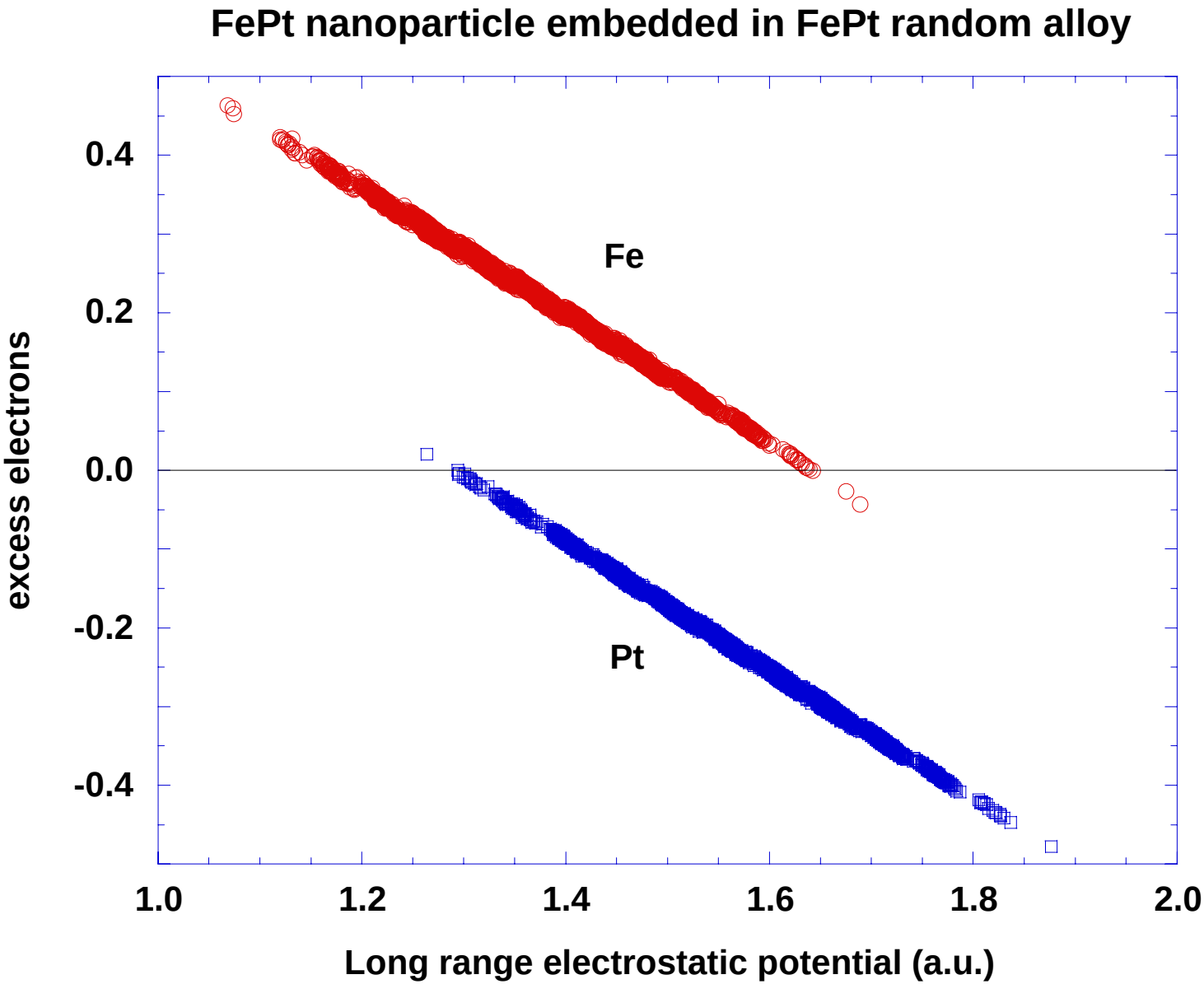
By Greg Foss at PSC


FePt nanoparticle embedded in FePt random alloy
excess electrons
0.4
0.2
0.0
-0.2
-0.4
Fe
Fe
FePt (L1$_0$)
nanoparticle
(diameter = 38.6)
FePt (fct)
random alloy
Pt
Pt
0
10
20
30
40
50
60
distance from the center (Angstrom)


FePt nanoparticle embedded in FePt random alloy
magnetic moment (Bohr magneton)
3.2
2.4
1.6
0.8
0.0
0
10
20
30
40
50
60
distance from the center (Angstrom)
Fe
Fe
Pt
Pt
FePt (L1$_0$)
nanoparticle
(diameter = 38.6)
FePt (fct) random alloy


FePt nanoparticle embedded in FePt random alloy
excess electrons
0.4
0.2
0.0
-0.2
-0.4
0
2
4
6
8
10
12
number of unlike nearest neighbors
Fe
Pt
excess electrons on Fe
excess electrons on Pt


FePt nanoparticle embedded in FePt random alloy
magnetic moment (Bohr magneton)
number of unlike nearest neighbors
magnetic moment on Fe
magnetic moment on Pt
Fe
Pt
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
0
2
4
6
8
10
12


FePt nanoparticle embedded in FePt random alloy
excess electrons
0.4
0.2
0.0
-0.2
-0.4
Fe
Pt
1.0
1.2
1.4
1.6
1.8
2.0
Long range electrostatic potential (a.u.)

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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