Calculated Magnetic Structure in irradiated Fe

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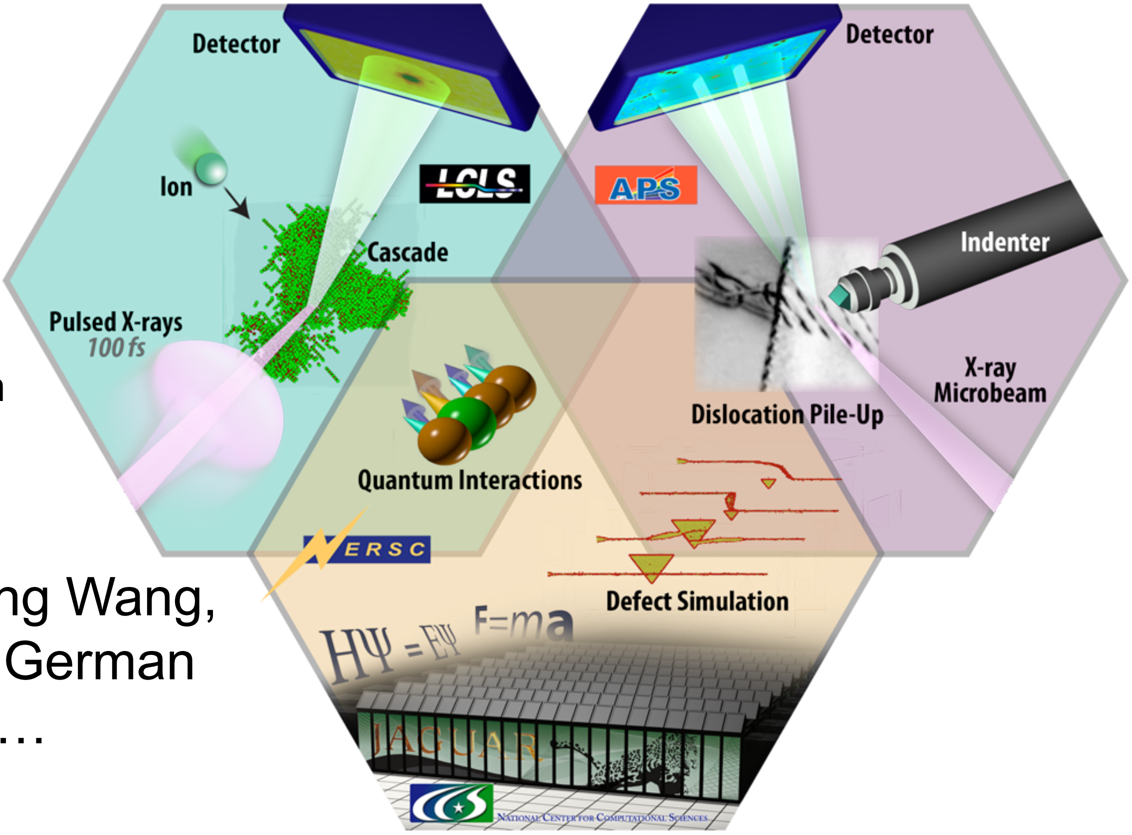
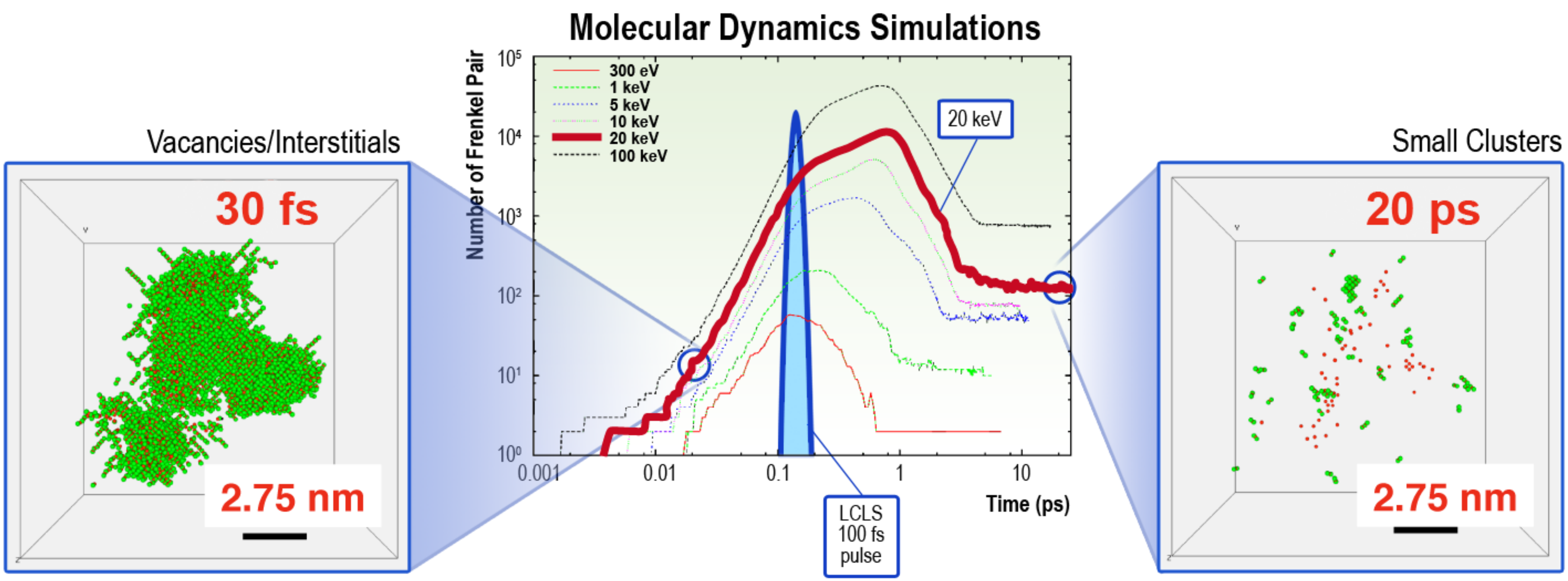


Thanks to: Malcolm Stocks, Yang Wang, Kh. Odbadrakh, Roger Stoller, German Samolyuk, Madhu Ojha…

CENTER FOR DEFECT PHYSICS (CDP)
an Energy Frontier Research Center EFRC

Defect Production During Irradiation



Primary damage formation

- Essentially everything that is known about primary damage formation comes from classical molecular dynamics (MD) simulations
- The first few picoseconds set the stage

Molecular dynamics simulations of displacement cascade evolution have spanned 50 years

Experimental measurements of cascade dynamics and evolution will be possible *for the first time* with sub-picosecond X-ray pulses

Diffusion Quantum Monte Carlo (Randy Hood…)

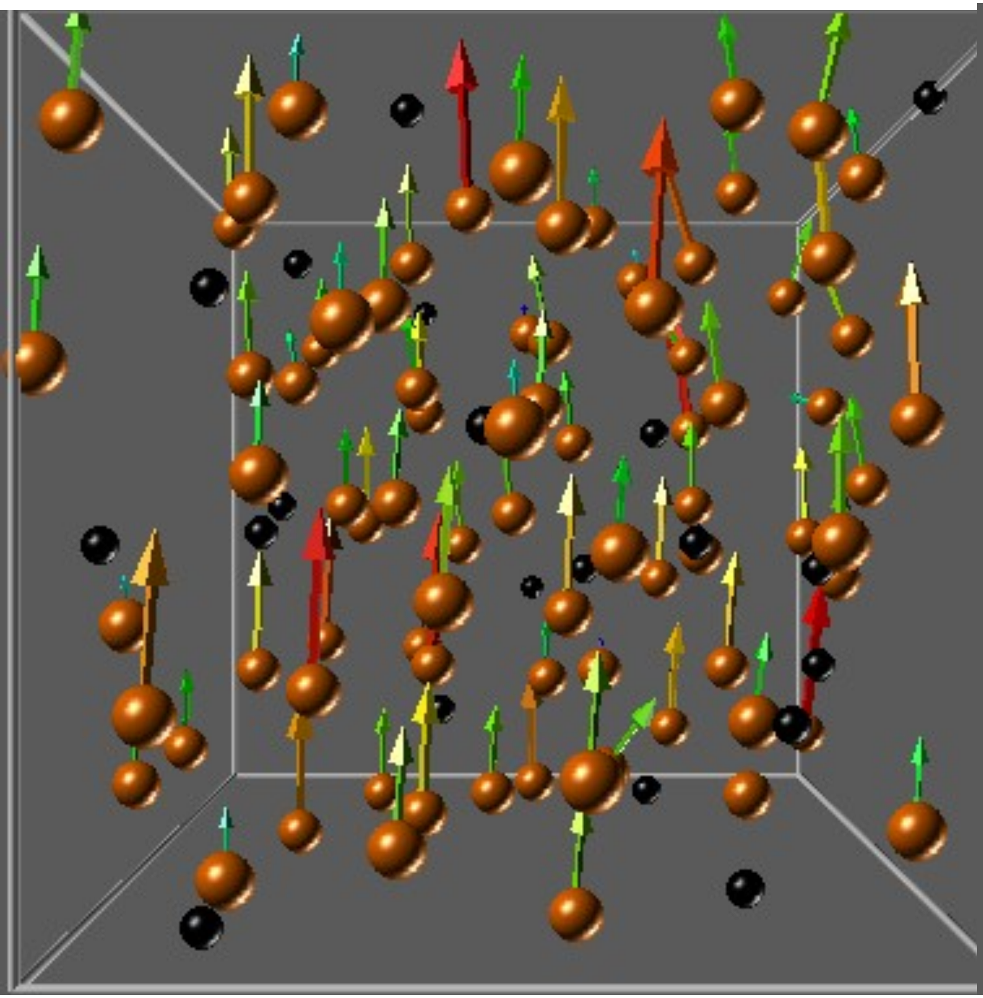
System	Method	Energy (eV)	Notes
Vacancy formation	DMC	0.668(8)	124 atom cell
	Expt	0.67(3), 0.67, 0.66(2)	
Vacancy migration	DMC	0.64(1)	124 atom cell
	Expt	0.62,0.61(3),0.65(6)	
NN divacancy binding	GGA	-0.07	123 atom cell
	DMC	-0.10(1)	123 atom cell
	Expt	"0.17(5)"	

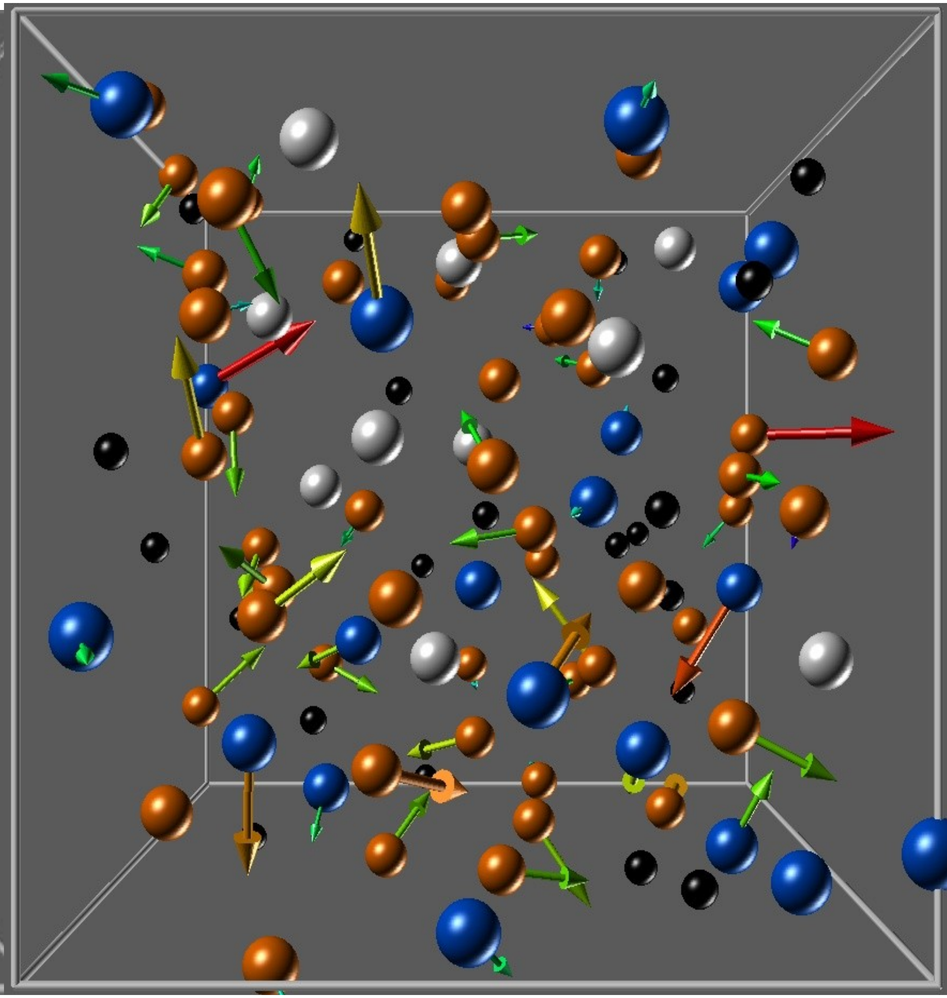
He substitutional formation	GGA	1.53	5^3 cell
	DMC	1.64(2)	4^3 cell 13^3 twists
He octahedral interstitial formation	GGA	3.24	5^3 cell
	DMC	3.53(1)	4^3 cell 13^3 twists
He tetrahedral interstitial formation	GGA	3.35	5^3 cell
	DMC	3.71(1)	4^3 cell 13^3 twists

12 MILLION HOURS!!!!!!!!!

CDP

Magnetic Structure [VASP/LSMS]

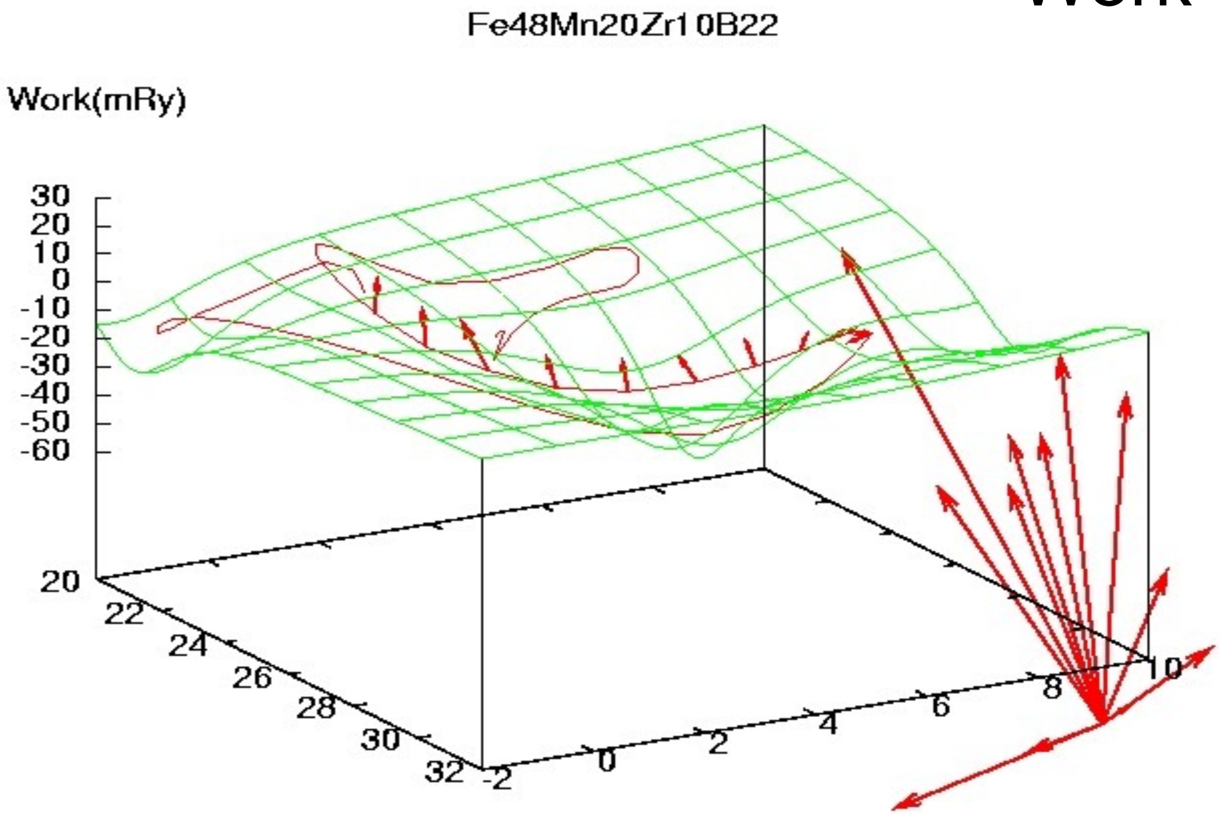




- $Fe_{.78}B_{.22}$
- $Fe_{.48}Mn_{.20}Zr_{.10}B_{.22}$

Diffusion in Fe48Mn20Zr10B22

Work= Fdx



Theory

DFT: $E_v^0 = Min_\rho E_v\left[\rho\right]$ $\qquad E_v\left[\rho\right] = \int \rho v + F\left[\rho\right]$

$$F\left[\rho\right] = Min_{\psi\to\rho}\left\langle \psi \middle| T + U \middle| \psi \right\rangle$$

Approximate DFT: $F\left[\rho\right] \approx T_s\left[\rho\right] + U_{classical}\left[\rho\right] + E_x\left[\rho\right] + E_c\left[\rho\right]$

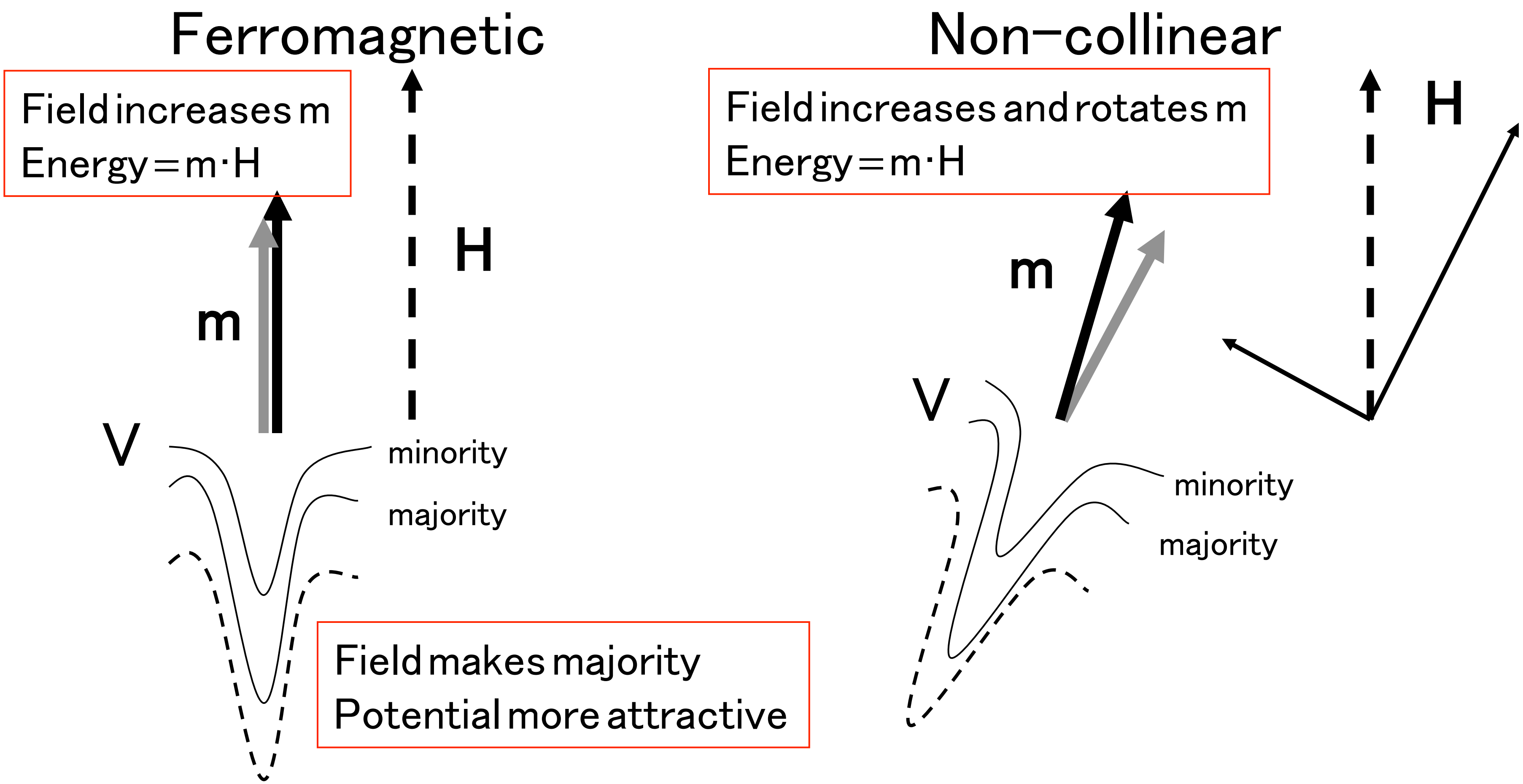
Other stuff: $E_v\left[\rho_A + \rho_B\right]$ $\qquad E_v\left[\rho_\uparrow + \rho_\downarrow\right]$ $\qquad E_v\left[\begin{pmatrix} \rho_{\uparrow\uparrow} & \rho_{\uparrow\downarrow} \\ \rho_{\downarrow\uparrow} & \rho_{\downarrow\downarrow} \end{pmatrix}\right]$

$$\begin{pmatrix} v_{\uparrow\uparrow} & v_{\uparrow\downarrow} \\ v_{\downarrow\uparrow} & v_{\downarrow\downarrow} \end{pmatrix} = v_0 I + v_x\sigma_x + v_y\sigma_y + v_z\sigma_z$$

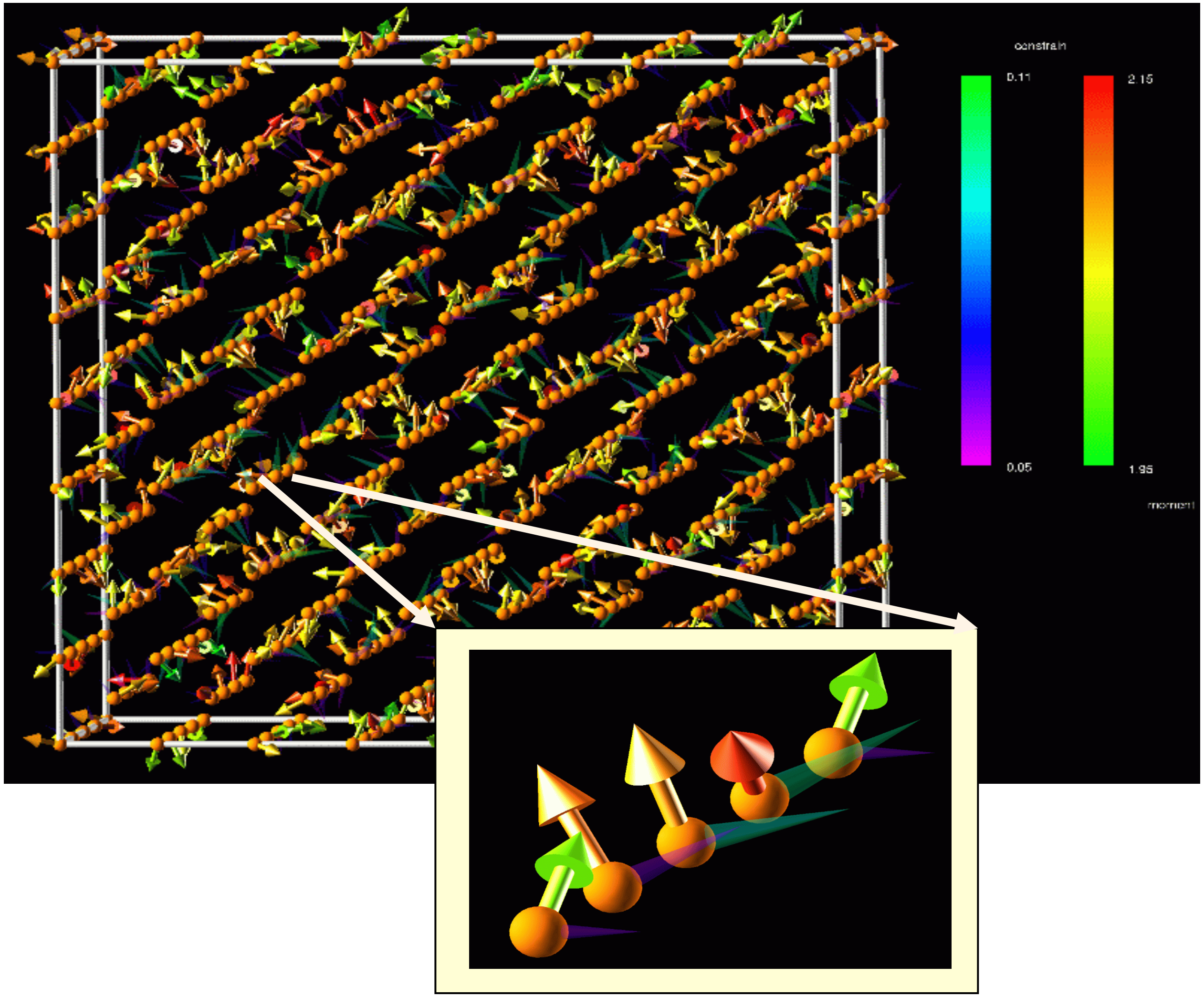
$$H = \begin{pmatrix} -\nabla^2 & 0 \\ 0 & -\nabla^2 \end{pmatrix} + \begin{pmatrix} v_\uparrow & 0 \\ 0 & v_\downarrow \end{pmatrix} \qquad H = \begin{pmatrix} -\nabla^2 & 0 \\ 0 & -\nabla^2 \end{pmatrix} + \begin{pmatrix} v_{\uparrow\uparrow} & v_{\uparrow\downarrow} \\ v_{\downarrow\uparrow} & v_{\downarrow\downarrow} \end{pmatrix}$$

$$L = \left\langle \psi \middle| i\hbar\frac{d}{dt} - H \middle| \psi \right\rangle \qquad L = \left\langle \psi_{\{\mathbf{m}_i\}} \middle| i\hbar\frac{d}{dt} - H \middle| \psi_{\{\mathbf{m}_i\}} \right\rangle \qquad \hbar\dot{\mathbf{m}} = \mathbf{m}\times\mathbf{B}$$

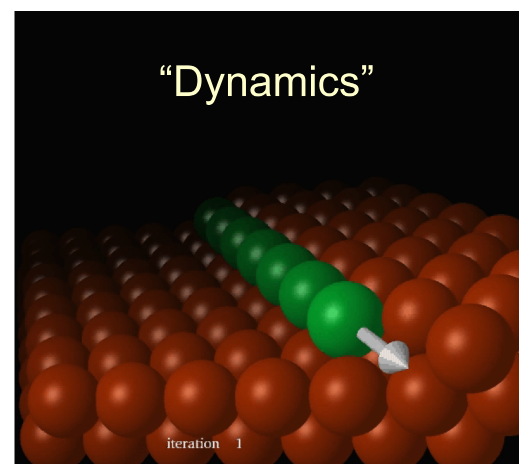
Application of Field - Schematic



Existence of CLM States: DLM Fe



- Constrained Local Moment States in Fe
  - View CLM state as a single time step in a first principles Spin Dynamics simulation
  - Model Paramagnetic Fe : *(Iron above its Curie temperature)*
  - *8x8x4 repeat of body centered cubic cell :- 512 sites*
  - *Randomly distribute orientations*

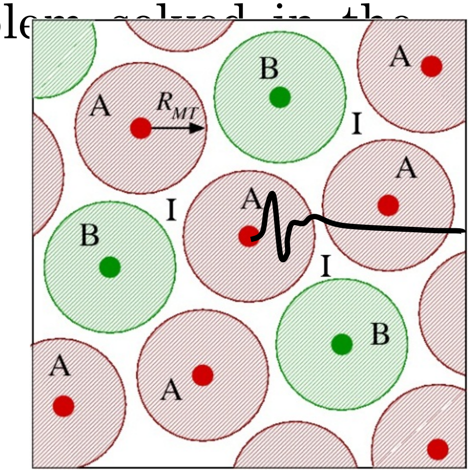


Co on Pt (Ujfalussy)

nuclear-electron interaction
all-electron methods

- **<u>All-electron methods</u>**:
  - ➢ Core, semi-core and valence electrons treated on same footing
  - ➢ Bare Coulomb interaction used

- **Muffin-tin orbitals** (solid-state): divide space into atomic spheres and interstitial regions. Atomic problem solved in the spheres, and matched to interstitial solutions.
  - ➢ **KKR – LMTO (Hankel functions)**
  - ➢ **APW – LAPW (Plane waves)**
  - ➢ **ASW (Spherical waves)**



- **Localized basis sets** (quantum chemistry):
  - ➢ **Gaussian-type orbitals (GTO)**
  - ➢ **Slater-type orbitals (STO)**
  - ➢ **Linear combination of atomic orbitals (LCAO)**

Green's Function

Free electron Green's function :

$$(-\nabla^2 - E)G_0(r,r',E) = \delta(r-r')$$

$$G_0(r,r',E) = \frac{1}{4\pi}\frac{e^{i\sqrt{E}|r-r'|}}{|r-r'|}$$

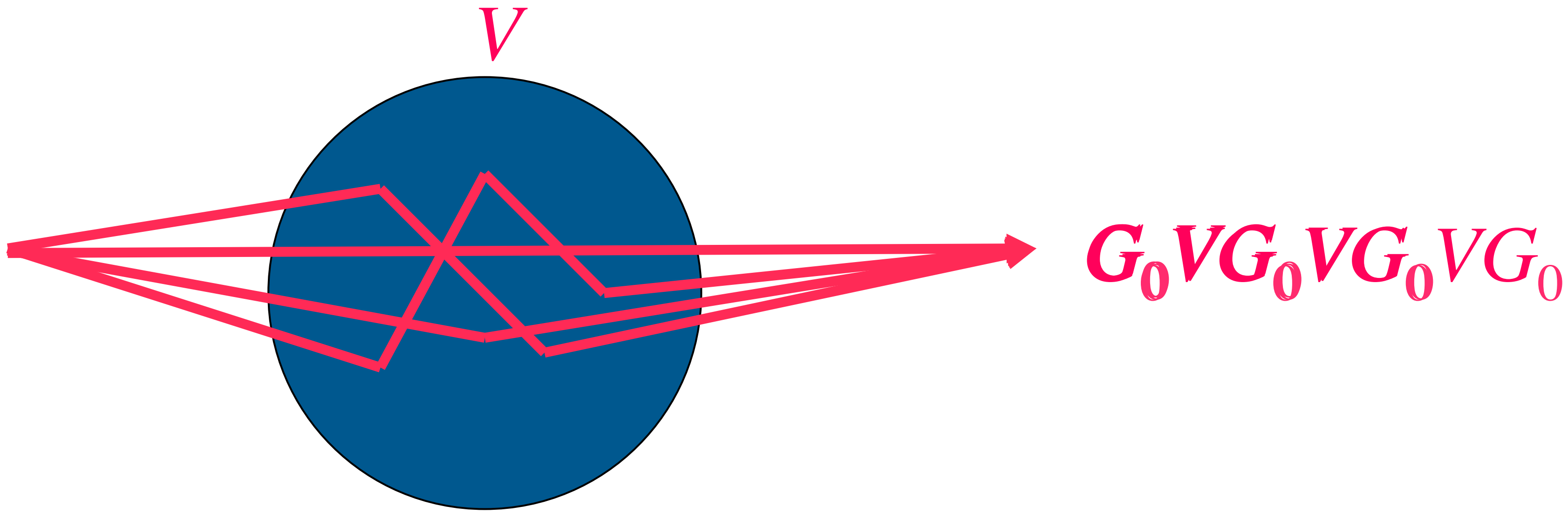
Wave function: $$\psi_k(\vec{r}) = \chi(\vec{r}) + \int G_0(\vec{r},\vec{r}')V(\vec{r}')\psi(\vec{r}')d^3\vec{r}'$$

$$G(\mathbf{r},\mathbf{r}',E) = G_o(\mathbf{r},\mathbf{r}',E) + \int G_o(\mathbf{r},\mathbf{r}'',E)\,V(\mathbf{r}'')\,G(\mathbf{r}'',\mathbf{r}',E)\,d\mathbf{r}''$$

$$n(\vec{r}) = -\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{E_F} TrG(\vec{r},\vec{r};\varepsilon)d\varepsilon$$

$$\vec{m}(\vec{r}) = -\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{E_F} TrG(\vec{r},\vec{r};\varepsilon)\vec{\sigma}d\varepsilon$$

Scattering at a single potential

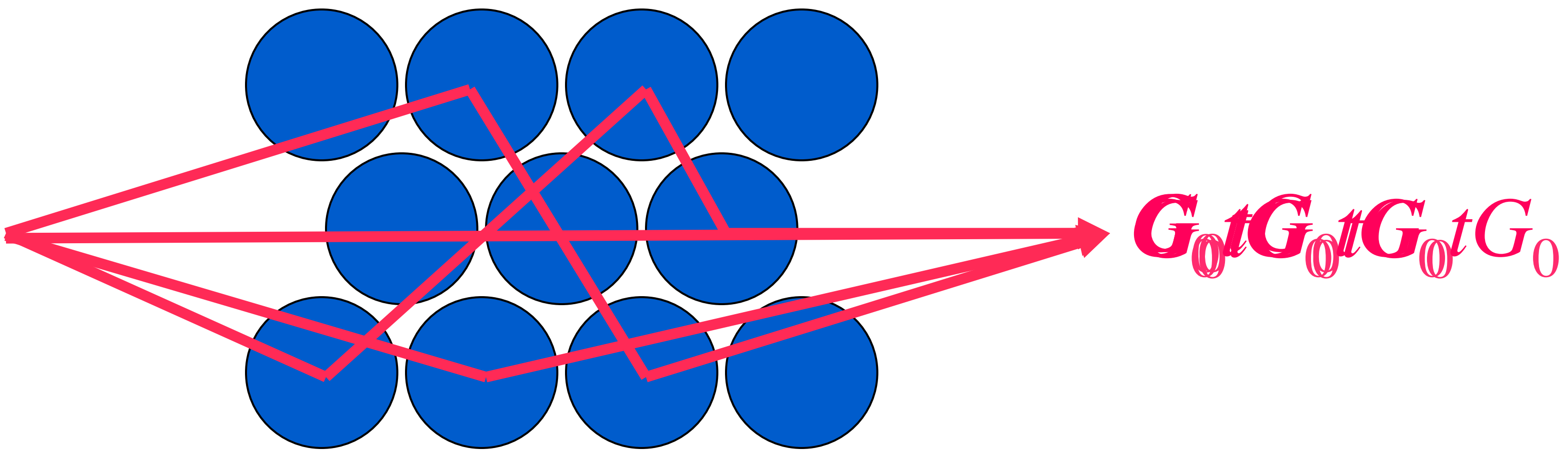


$$G = G_0 + G_0VG_0 + G_0VG_0VG_0 + G_0VG_0VG_0VG_0 + \ldots\ldots\ldots$$

$$= G_0 + G_0(V + VG_0V + VG_0VG_0V + \ldots\ldots\ldots)G_0$$

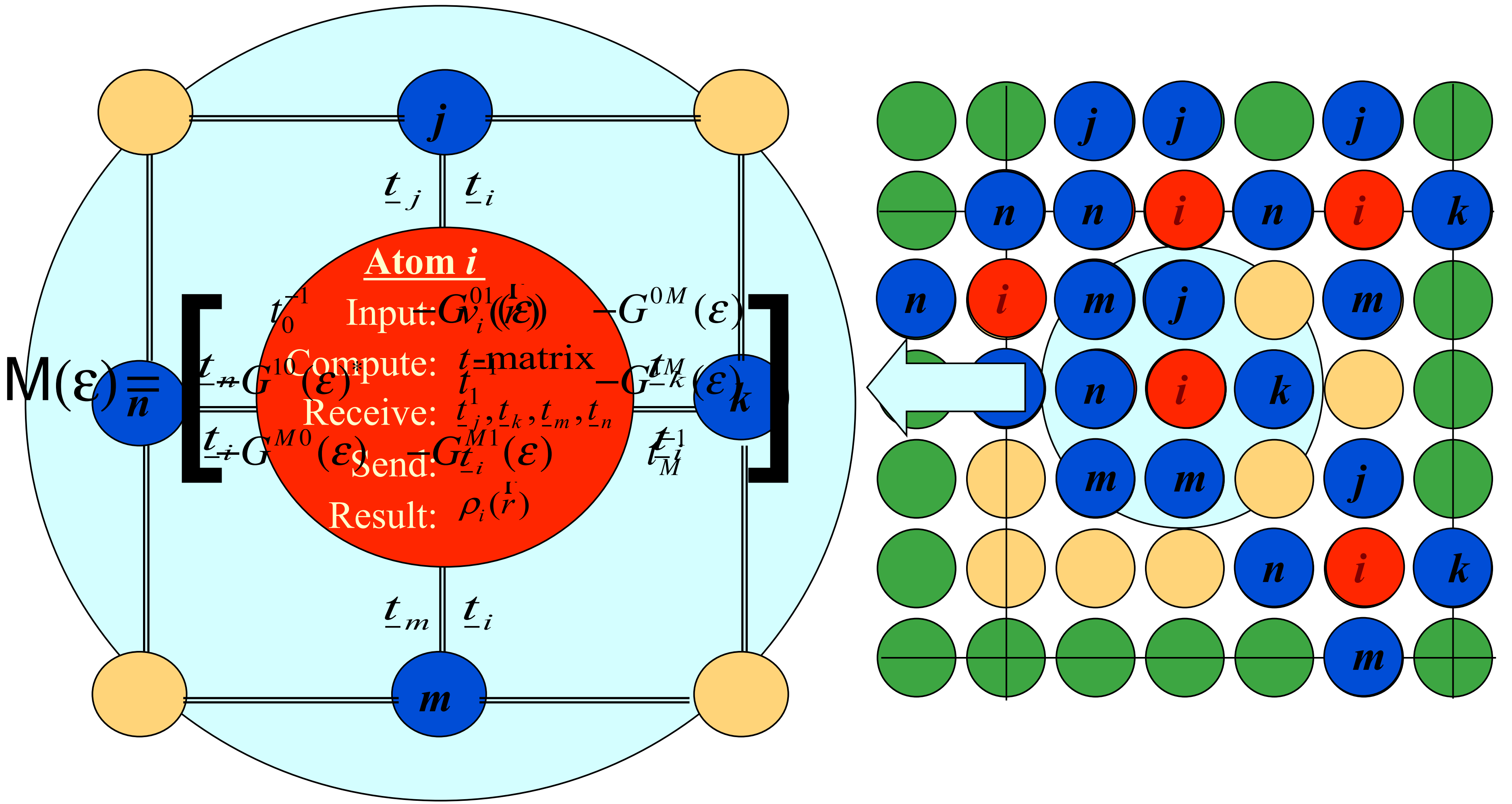
$$= G_0 + G_0tG_0$$

Multiple scattering



$$
\begin{aligned}
G &= G_0 + G_0 t G_0 + G_0 t G_0 t G_0 + G_0 t G_0 t G t G_0 + \ldots\ldots \\
&= \mathrm{G}_0 + G_0 T G_0 \\
&= \frac{G_0}{1 - t G_0}
\end{aligned}
$$

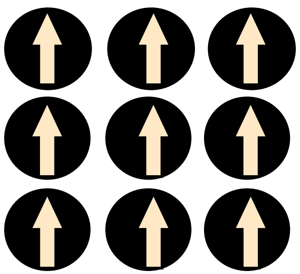
O[N] electronic Structure Methods for Extended Defects



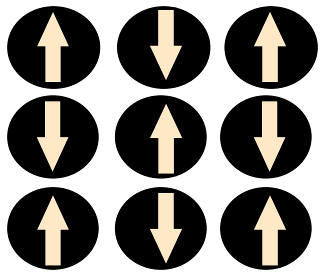
LSDA and Finite Temperature Magnetism

- **LSDA is theory of ground states**

FM

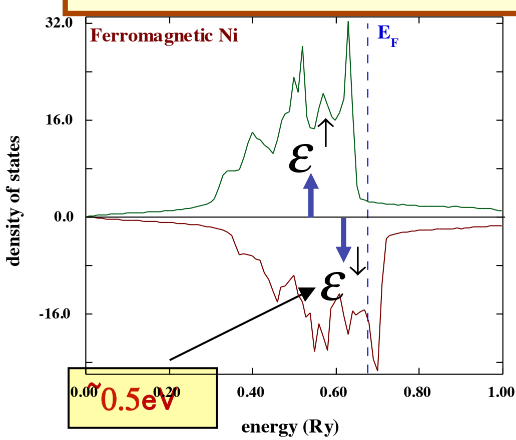


AFM



- **Correct magnetic ground state**

Moment (m_B)	Fe	Ni
Calc. LSDA	2.17	0.59
Expt.	2.08	0.57



Stoner like:

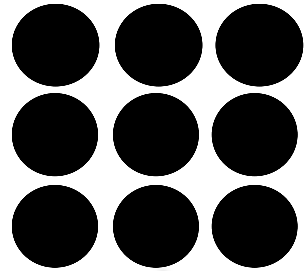
$$M = I\Delta_{Exch}$$

$$\Delta_{Exch} \sim \varepsilon^{\uparrow} - \varepsilon^{\downarrow}$$

$$I \sim 1.0\mu_B Ry^{-1}$$

- **LSDA magnetism at finite temperature**

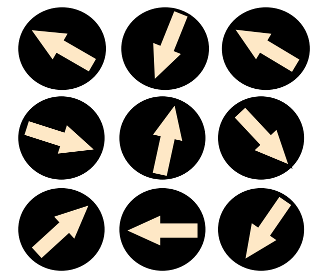
NM



T_C (K)	Fe	Ni
Stoner	20,000	10,000
Expt.	1040	640

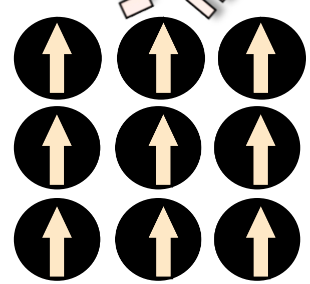
$$T_C^{Stoner} \sim M / I$$

PM
Disordered Local Moment (DLM) state



$$T_C^{DLM} = T_C^{Stoner}$$

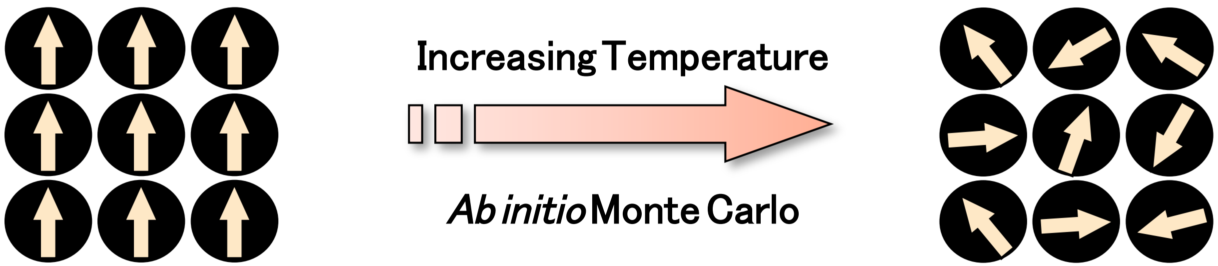
T_C (K)	Fe	Ni
DLM	1015	450
Expt.	1040	640



T=0K; FM

Finite Temperature Magnetism: Locally Selfconsistent Multiple Scatering (LSMS), Order N constrained DFT

- **Statistical Physics of Moment Orientations**



➢ **Wang-Landau Monte Carlo algorithm and high performance computing facilitate *ab initio* studies of finite temperature magnetic fluctuations**

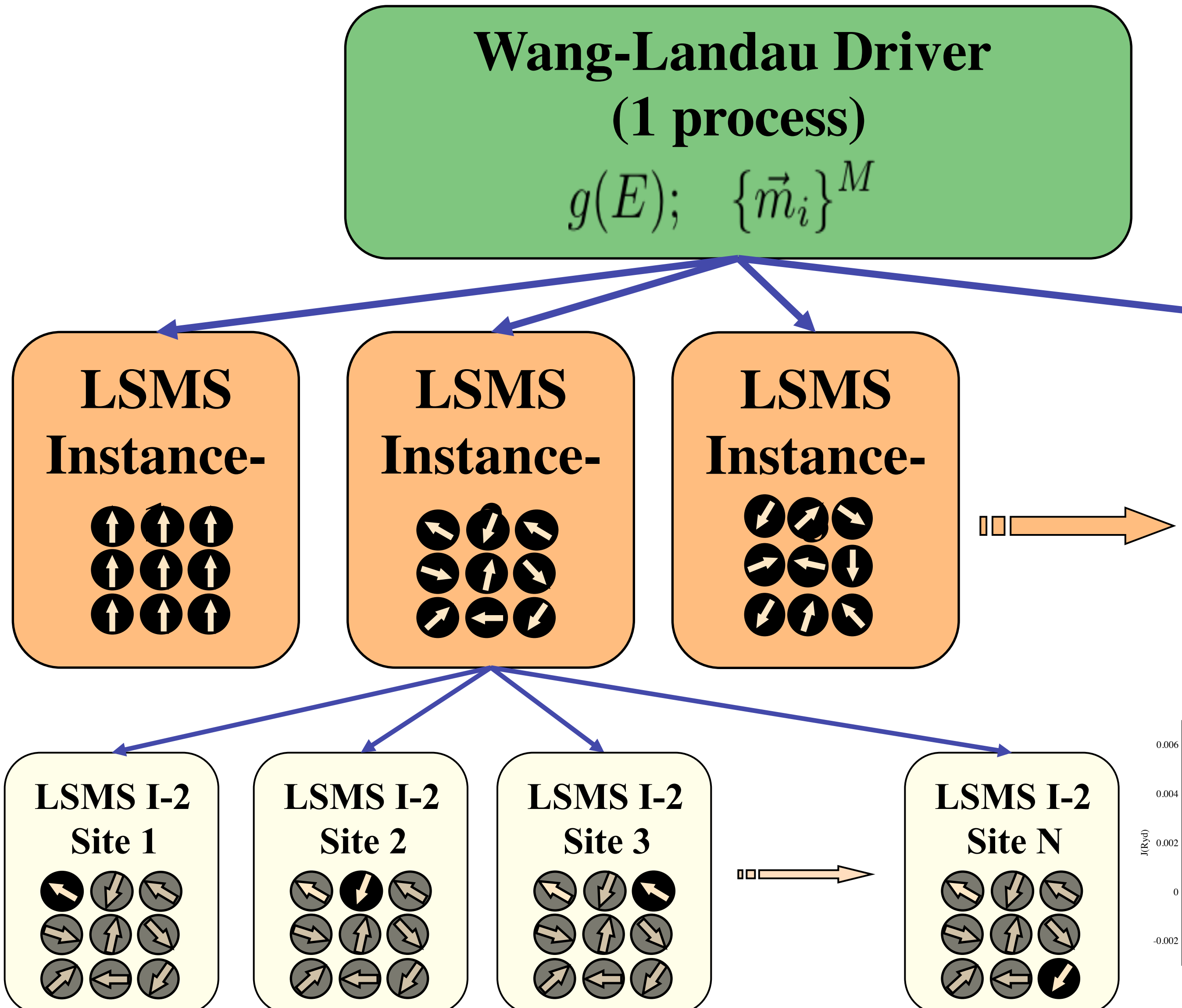
- **Calculate statistical density of states**
- **Thermodynamics at all temperatures**

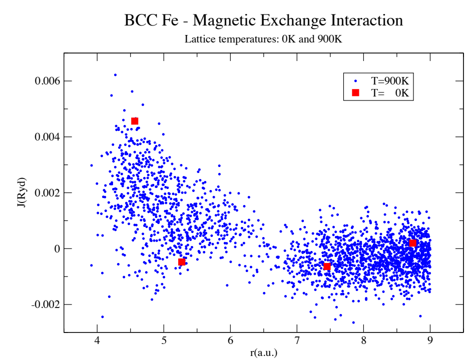
$$Z = \int g(E) e^{-E/k_B T} dE$$

$$\Delta G(T) = \int_0^{H_{\max}} M(T)\, dH$$

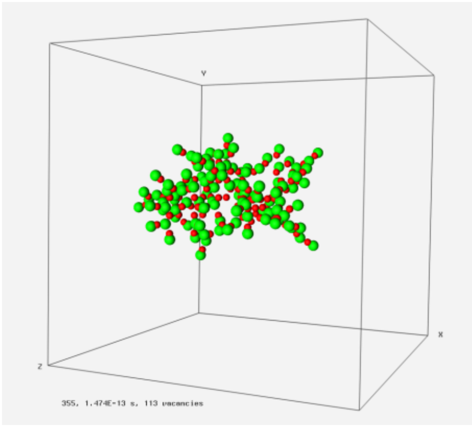
Cray-Titan peak speed 10 - 20 petaflops. Latest AMD Opteron and NVIDIA Tesla, 299,008 cores/ 600 terabytes of memory

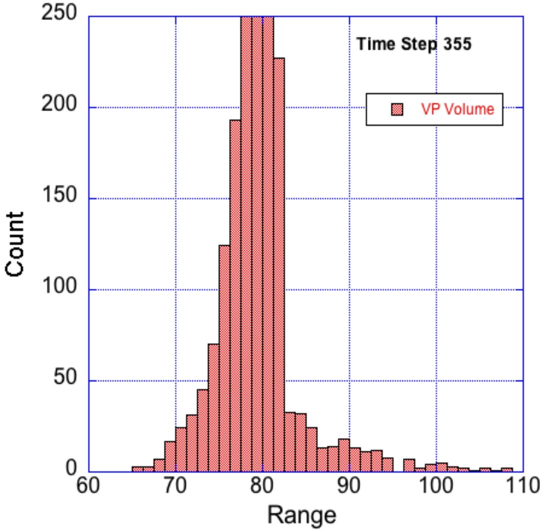
Wang-Landau-LSMS allows multi-level parallelism

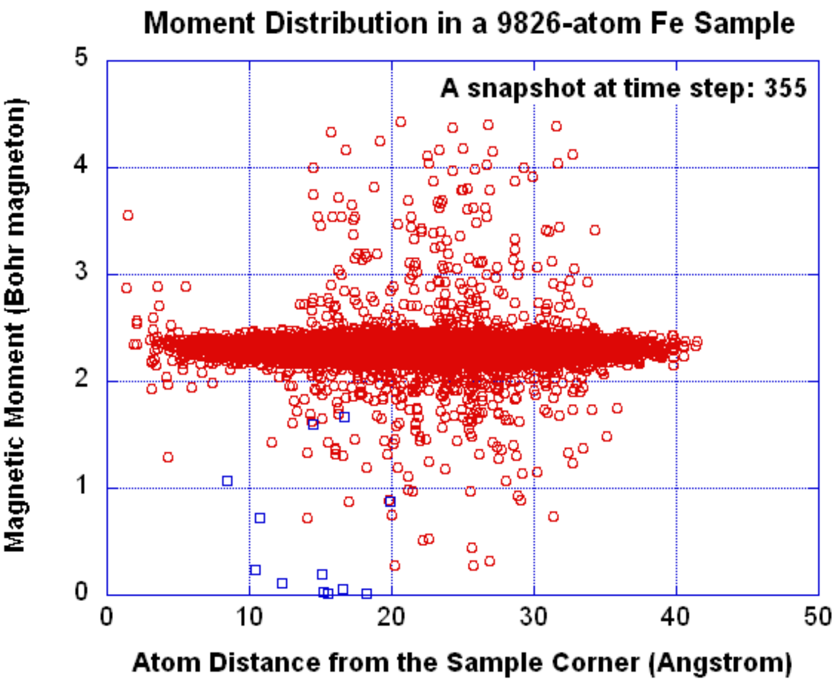


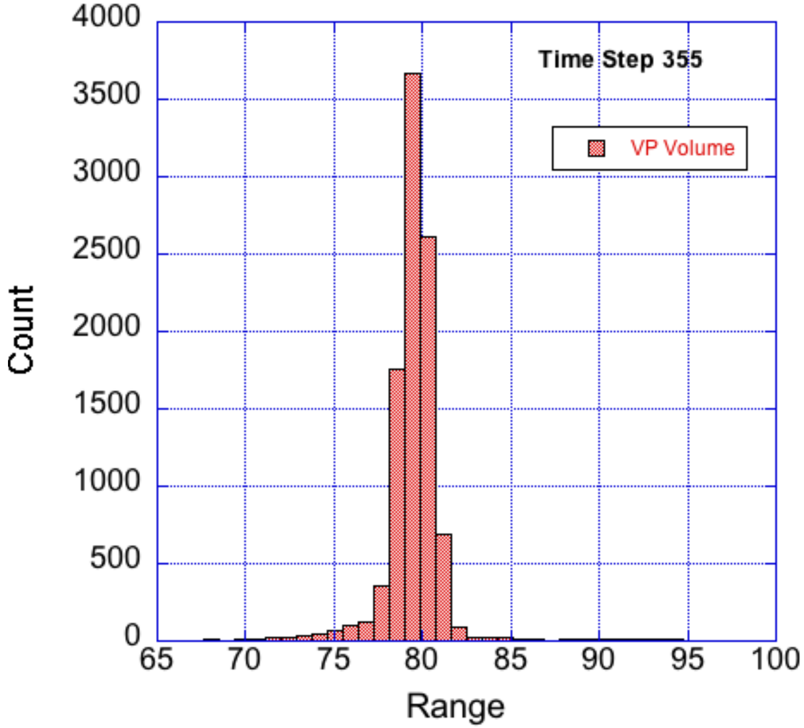


Ab Initio Simulations of Magnetic State

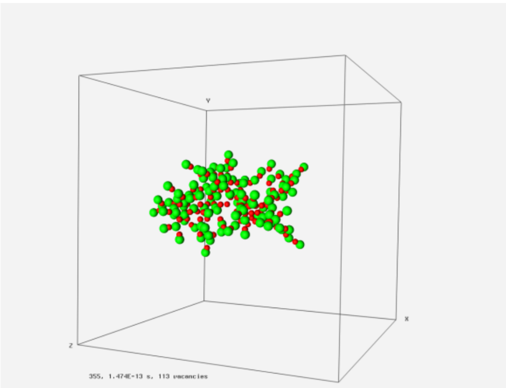
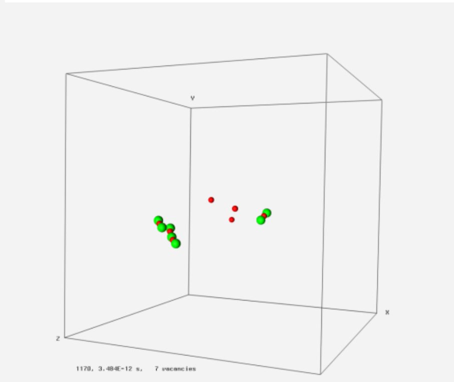
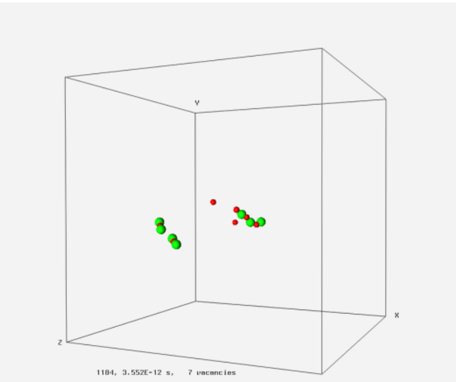
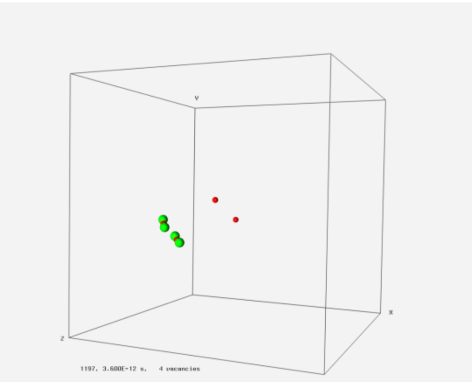


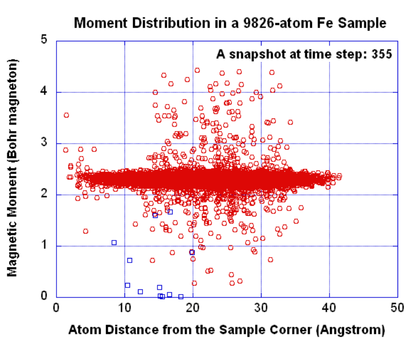


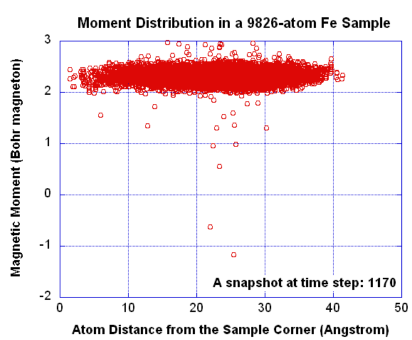


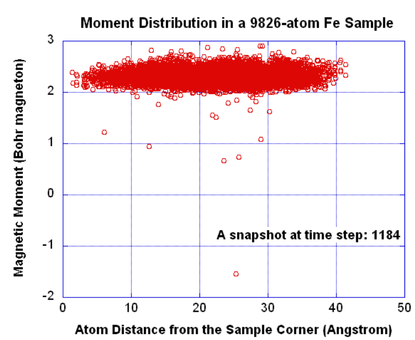


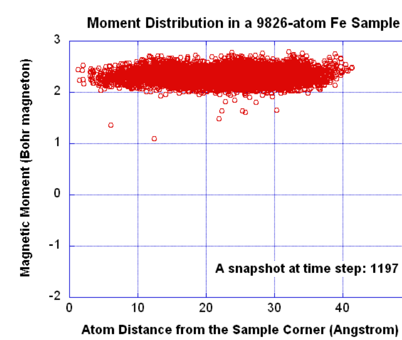
Ab Initio Simulations of Magnetic State

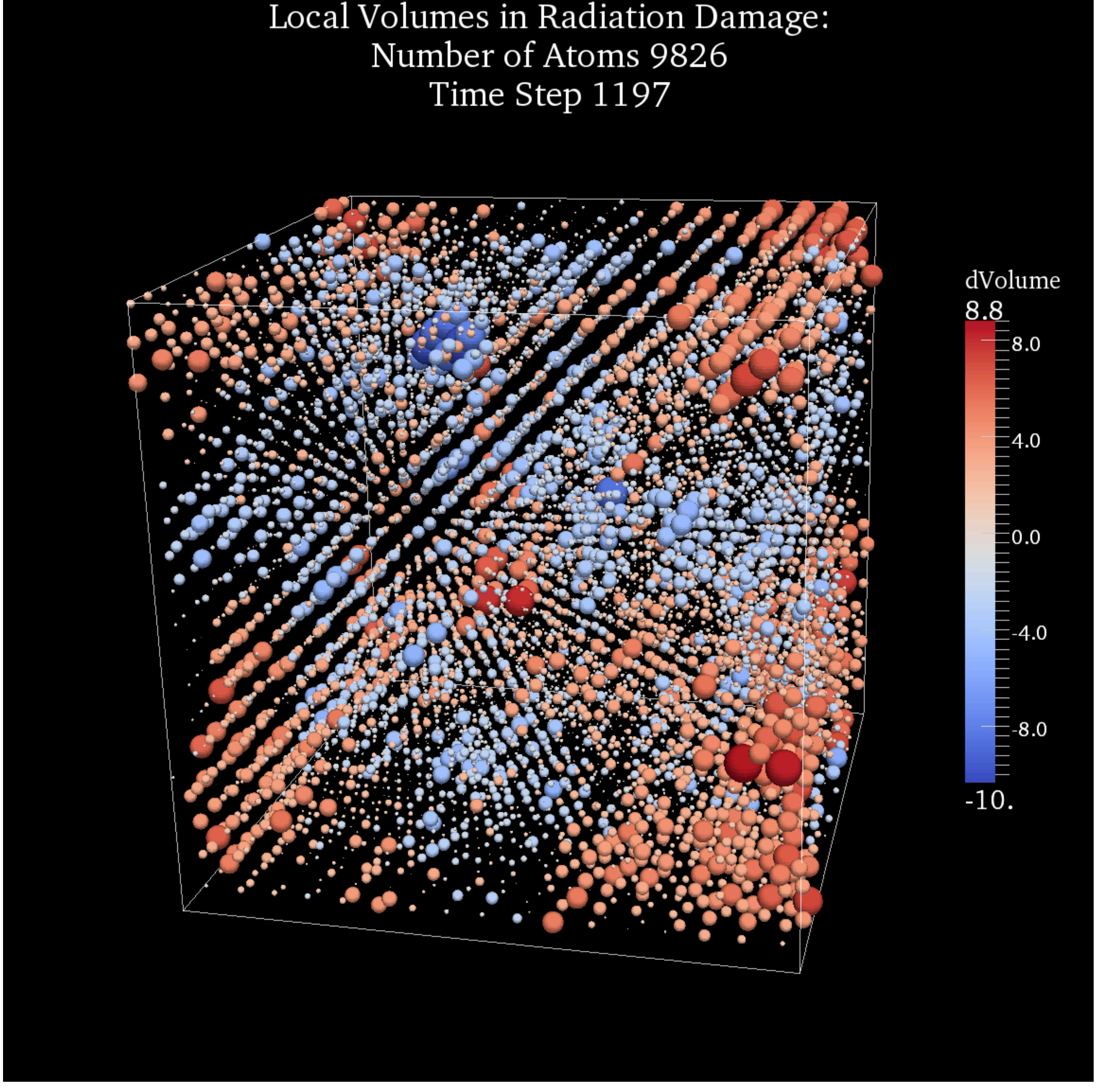





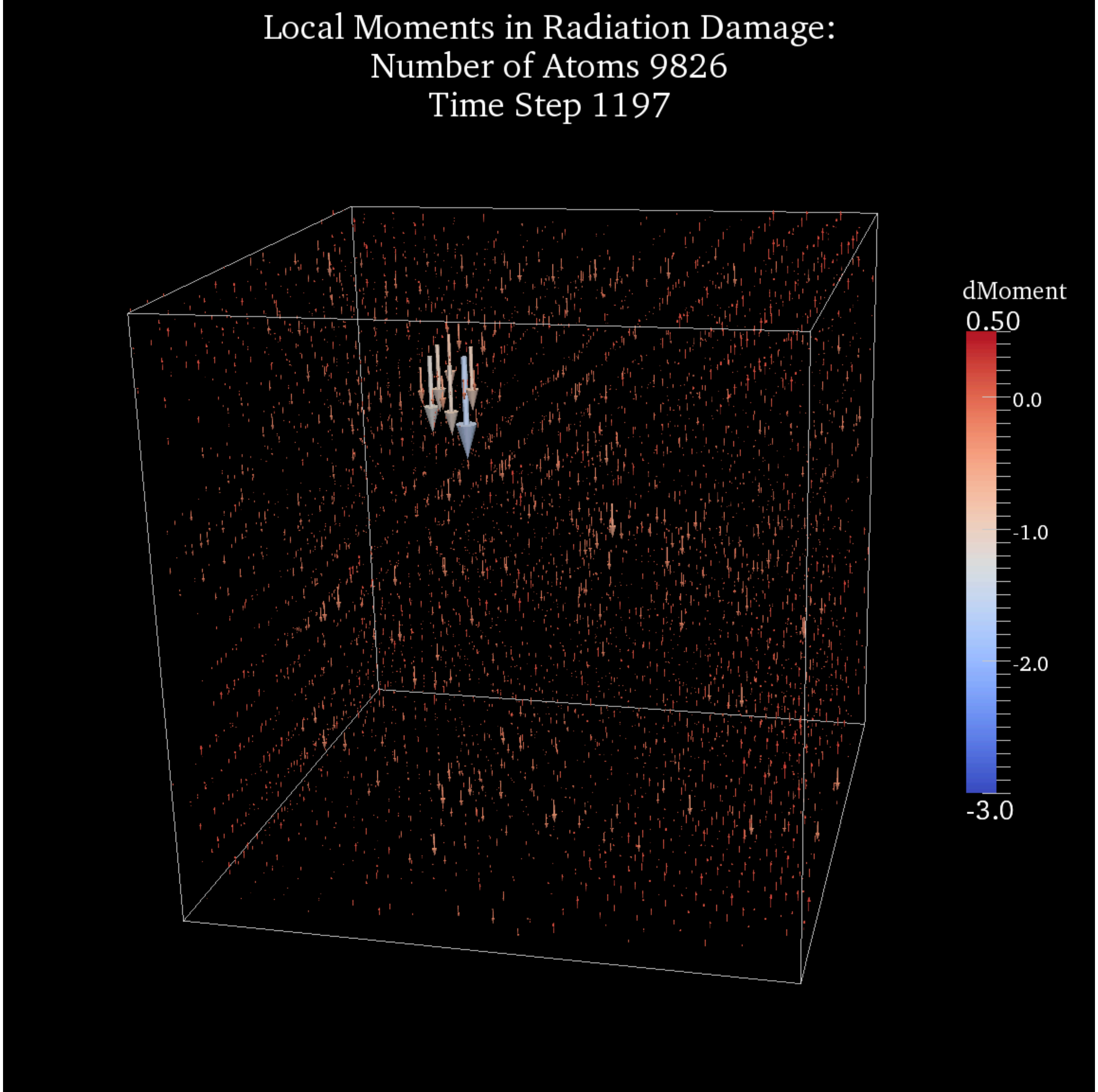


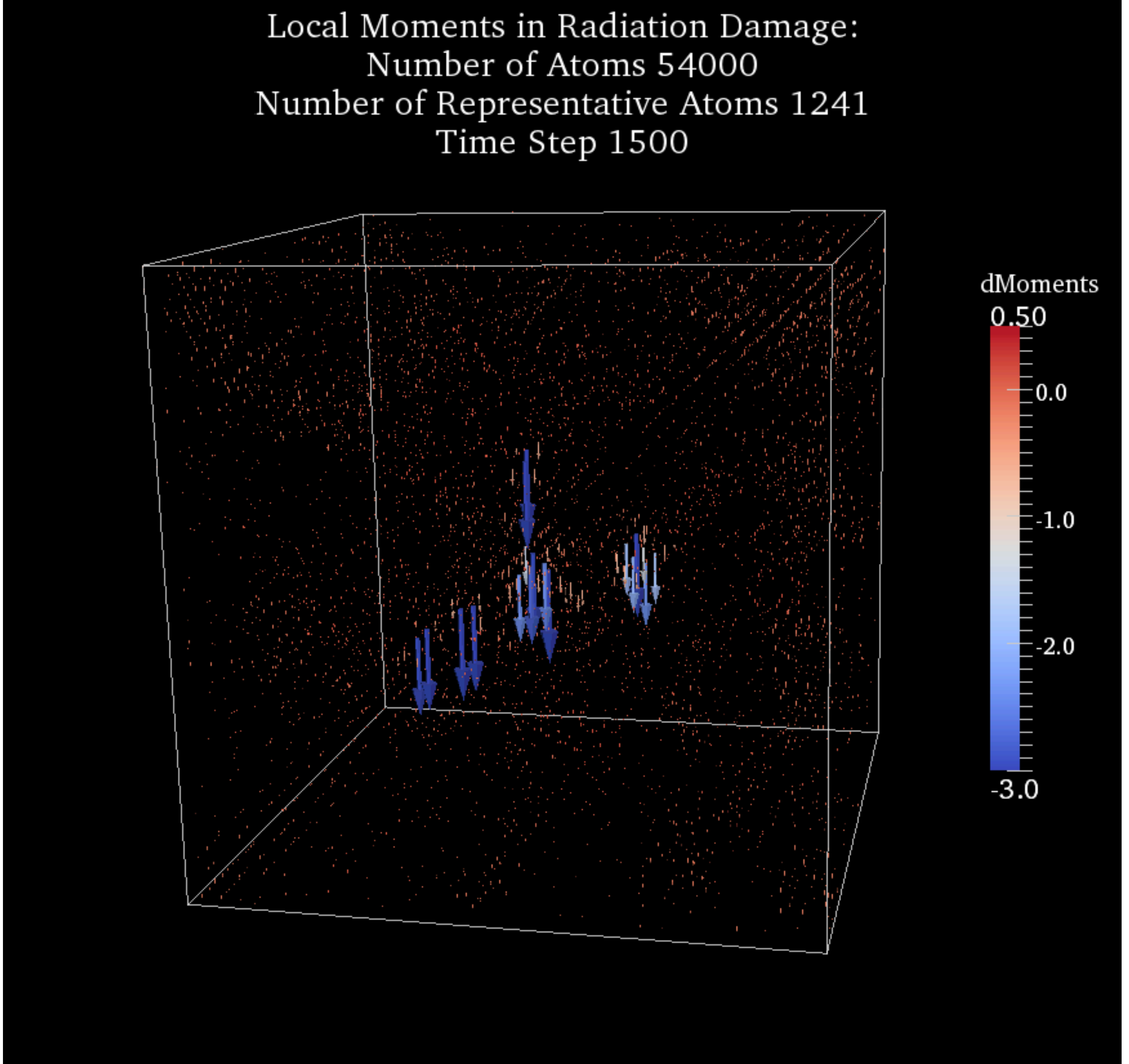







Local Volumes in Radiation Damage:
Number of Atoms 9826
Time Step 1197
dVolume
8.8
8.0
4.0
0.0
-4.0
-8.0
-10.


Local Moments in Radiation Damage:
Number of Atoms 9826
Time Step 1197
dMoment
0.50
0.0
-1.0
-2.0
-3.0


Local Moments in Radiation Damage:
Number of Atoms 54000
Number of Representative Atoms 1241
Time Step 1500
dMoments
0.50
0.0
-1.0
-2.0
-3.0

Summary

- The study of defects pushes us to develop techniques for the arbitrary, large, complex structures that are important for structural materials, information processing, energy collection, biology…

- There are many challenges: formalism, algorithms, computer science, hardware.

- The study of unit defect events pushes experiment toward the small and fast and theory toward the large and slow.

Acknowledgment

This research was performed at Oak Ridge National Laboratory (ORNL) and is based upon work supported as part of the Center for Defect Physics in Structural Materials (CDP), an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences. This research used resources of the Oak Ridge Leadership Computing Facility at ORNL, which is supported by the Office of Science of the Department of Energy under contract DE-AC05-00OR22725.