Computing the Magnetic Structure at Interfaces

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Magnetic Nanostructures, Interfaces, and New Materials
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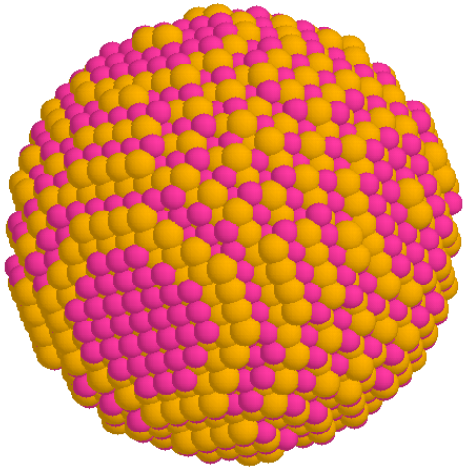
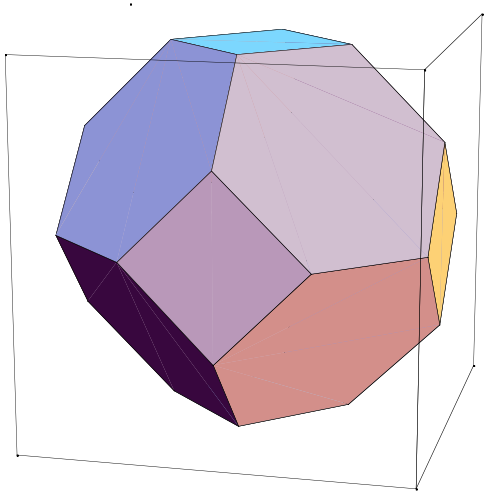
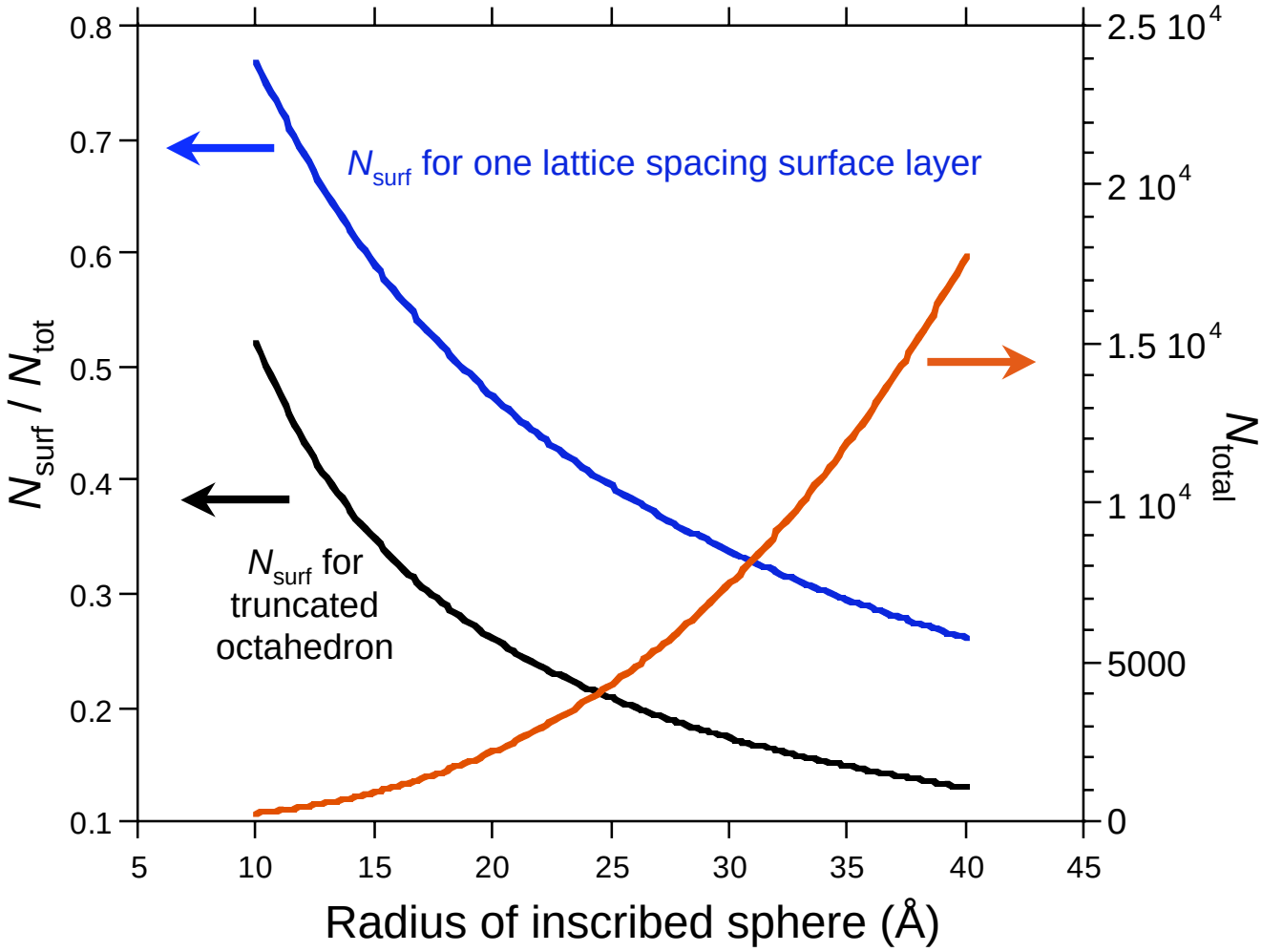
- **Outline:**
  - **Magnetic nanostructures: opportunities & challenges**
  - ***Ab initio* modeling of magnetic systems**
  - **FeMn and FeMn|Co heterostructures**
  - **Future plans**

- **Collaborators:**
  - **Balazs Ujfalussy (ORNL & Budapest, Hungary)**
  - **Malcolm Stocks (M&C Division, ORNL)**

Example: FePt (L10) Nanoparticle







⇒ Nanoscale is dominated by interface

Surface Spin Disorder in Nanoparticles

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Surface Spin Disorder in $NiFe_2O_4$ Nanoparticles

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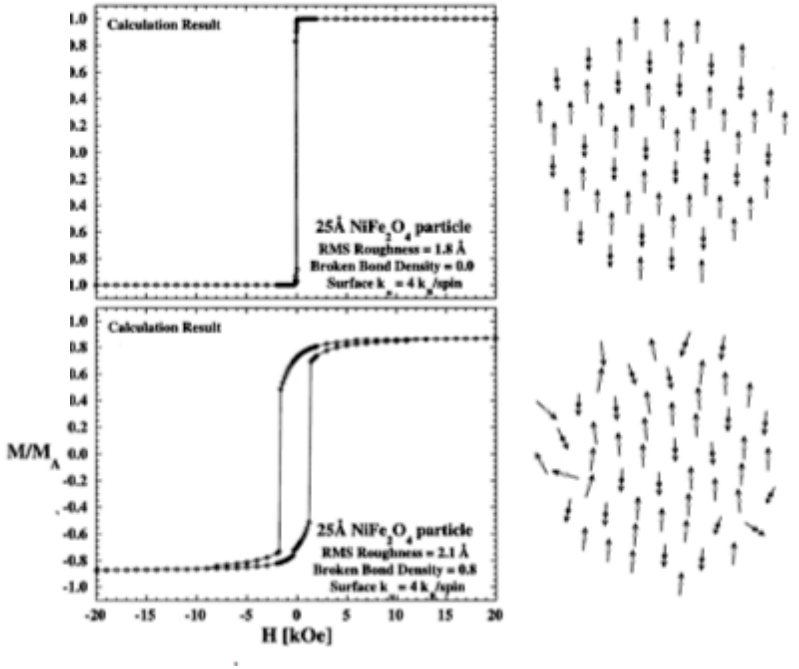
(Received 6 February 1996)

Nickel ferrite nanoparticles exhibit anomalous magnetic properties at low temperatures: low magnetization with a large differential susceptibility at high fields, hysteresis loops which are open up to 160 kOe, time-dependent magnetization in 70 kOe applied field, and shifted hysteresis loops after field cooling. We propose a model of the magnetization within these particles consisting of ferrimagnetically aligned core spins and a spin-glass-like surface layer. We find that qualitative features of this model are reproduced by a numerical calculation of the spin distribution. Implications of this model for possible macroscopic quantum tunneling in these materials are discussed. [S0031-9007(96)00628-X]

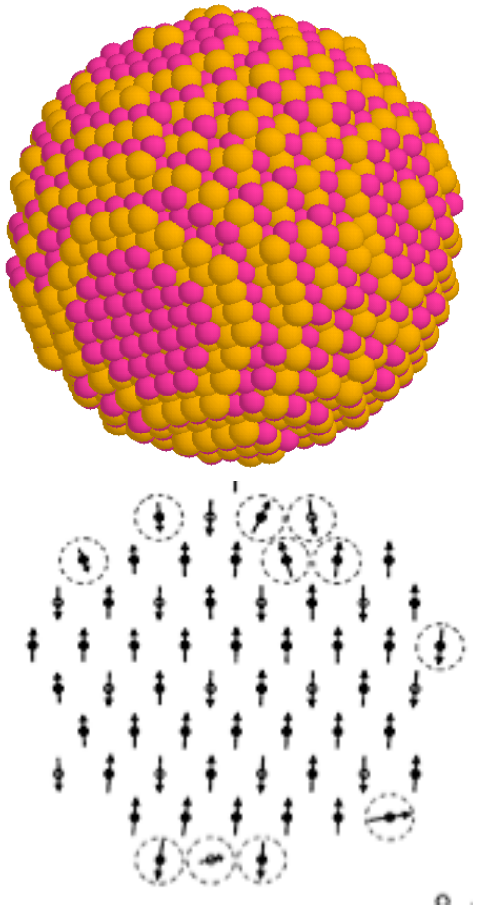
PACS numbers: 75.50.Tt, 75.30.Pd, 75.50.Gg, 75.50.Lk



- **Competition between surface anisotropy and exchange**
- **Maybe even random exchange**
- **Ferro- (Ferri-) magnetic interior couples to spin-glass (antiferromagnetic) surface region**
- **Enhancement of "effective" anisotropy**

Nanoparticles / Nanostructures

- Nano-particles/gains = “single domain”
- 6 nm particle ~40% of atoms in surface region
- Competition between chemical phases
  - Multiple magnetic phases
  - Surface segregation
- Demonstrated with Kodama’s work on $NiFe_2O_4$: magnetic structure probably complex!
- Interfaces between “materials” or magnetic phases
  - Ferro/antiferro; ferro/sin-glass; …
  - Exchange bias was discovered in Co nanopaticles!
- Because of large surface to volume ratio all interesting effects occur in one particle
- Interest / opportunities in magnetic systems is due to there complexity
  - Long range interaction; non-linearity of LLG; …



Complex spin structure in nanostructures could be an opportunity

Atomic scale spin-structure

- **Nanomagnetism, opportunities & challenges**
  - **need to understand atomic scale spin structure**
- **Experimental probes**
  - **Most average over 10-100 or more nanometers**
  - **Atomic scale spin-structure accessible by inference using models**
- **Models of magnetism at the nano scale**
  - **Surface / interface region is like new material**
  - **Parameters (exchange, anisotropy, etc.) unknown**

⇒ Complement experiment with first principle electronic structure calculations

Ab initio modeling of magnetism

Self Consistent Field Equations

$$\tilde{V}^{eff}[n,\mathbf{m}] = V^{ext}\tilde{1} + \mathbf{B}^{ext}\cdot\tilde{\sigma} + -\tilde{1}\int d\mathbf{r}' n(\mathbf{r})/|\mathbf{r}-\mathbf{r}'| + \tilde{1}\left(\frac{\delta\Omega_{xc}}{\delta n(\mathbf{r})} + \frac{\delta\Omega_{xc}}{\delta \mathbf{m}(\mathbf{r})}\right)\cdot\tilde{\sigma}$$

LSDA : exchange/correlation

$$\left\{\left(\varepsilon + \frac{\hbar}{2m}\nabla^2\right)\tilde{1} - \tilde{V}^{eff}\right\}\tilde{G}(\mathbf{r},\mathbf{r}';\varepsilon) = \tilde{1}\delta(\mathbf{r}-\mathbf{r}')$$

$$n(\varepsilon) = \mathrm{Im}\frac{1}{\pi}\int d\mathbf{r}\ \mathrm{Tr}\tilde{G}(\mathbf{r},\mathbf{r}';\varepsilon)$$

$$n(\mathbf{r}) = \mathrm{Im}\frac{1}{\pi}\int d\varepsilon\, f(\varepsilon-\mu)\ \mathrm{Tr}\tilde{G}(\mathbf{r},\mathbf{r}';\varepsilon)$$

$$\mathbf{m}(\mathbf{r}) = \mathrm{Im}\frac{1}{\pi}\int d\varepsilon\, f(\varepsilon-\mu)\ \tilde{\sigma}\mathrm{Tr}\tilde{G}(\mathbf{r},\mathbf{r}';\varepsilon)$$

$$\Omega = \int d\varepsilon\, f(\varepsilon-\mu)\varepsilon n(\varepsilon) - \frac{e^2}{2}\iint d\mathbf{r}d\mathbf{r}' n(\mathbf{r})n(\mathbf{r}')/|\mathbf{r}-\mathbf{r}'| + \Omega_{xc} - \int d\mathbf{r}\left\{\frac{\delta\Omega_{xc}}{\delta n(\mathbf{r})}n(\mathbf{r}) + \frac{\delta\Omega_{xc}}{\delta\mathbf{m}(\mathbf{r})}\cdot\mathbf{m}(\mathbf{r})\right\}$$

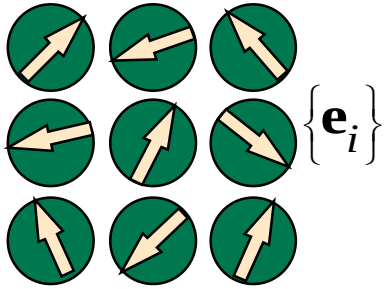
- **Magnetic moments:**
  - **Fe: ~2.3 μ_B exp. 2.2 μ_B**
  - **Co: ~1.6 μ_B exp. 1.7 μ_B**
  - **works for many other materials (Ni, Cr, etc.)**
- **Anisotropy: similar success**
- **Curie temperatures:**
  - **Mean Field with Onsager Cavity Fields**

	Fe	Ni
T_c (K)	1015	450
Expt.	1040	631
m_{DLM} (μ_B)	1.89	0.0

Gyorffy ,et al. J. Phys. F15, 1337 (1985); ibid 1761
Staunton & Gyorffy ,PRL 69, 371 (1992)

Constrained Local Moment Method

- **Local moment: (even in itinerant transition metals)**



$$\mathbf{e}_i = \frac{\int_{\Omega_i} d\mathbf{r}\ \mathbf{m}_M^i(\mathbf{r})}{\left| \int_{\Omega_i} d\mathbf{r}\ \mathbf{m}_M^i(\mathbf{r}) \right|}$$

- **Time evolution of magnetic structure**

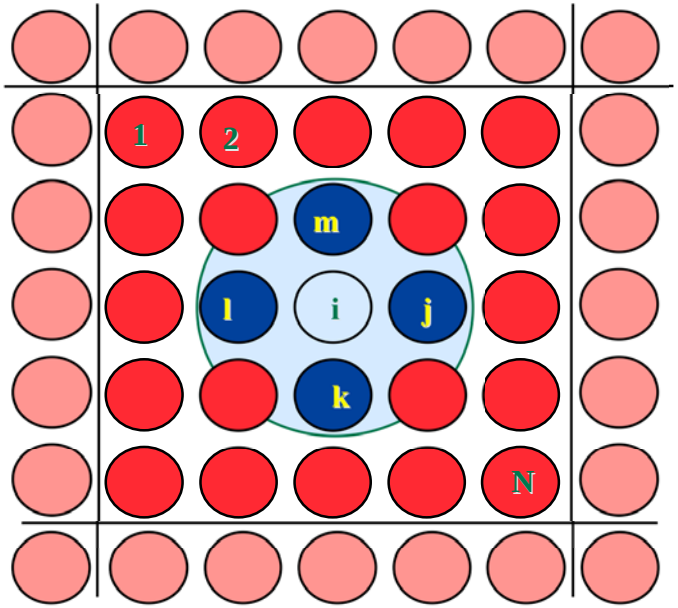
$$\frac{d}{dt}\mathbf{e}_i = -\frac{2\mu_B}{\hbar\bar{\mu}_i}[\mathbf{h}_i^{eff} \times \mathbf{e}_i]$$

$$\mathbf{h}_i^{eff} = \frac{\partial F}{\partial \mathbf{e}_i}$$

  - **Classical dynamics but ab initio free energy functional**

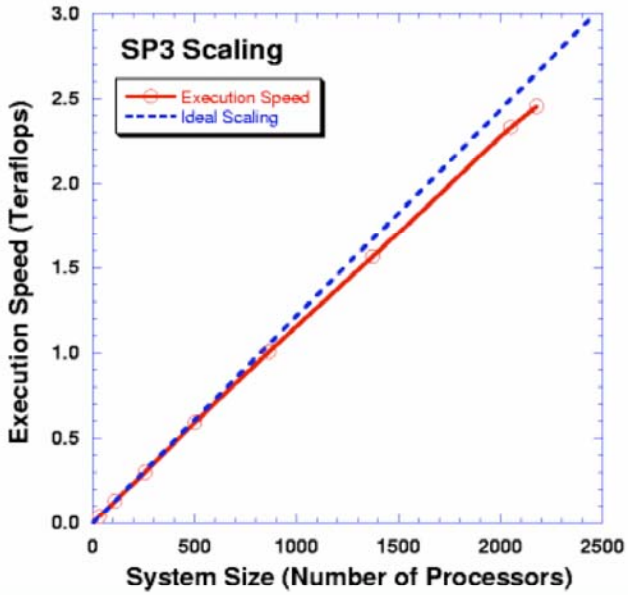
Locally Self-Consistent Method

- **Moment / charge of atom i**
  - **Schrödinger eq. in cluster**
  - **Poisson eq. in solid**



- **Algorithm maps perfectly onto massively parallel computers**

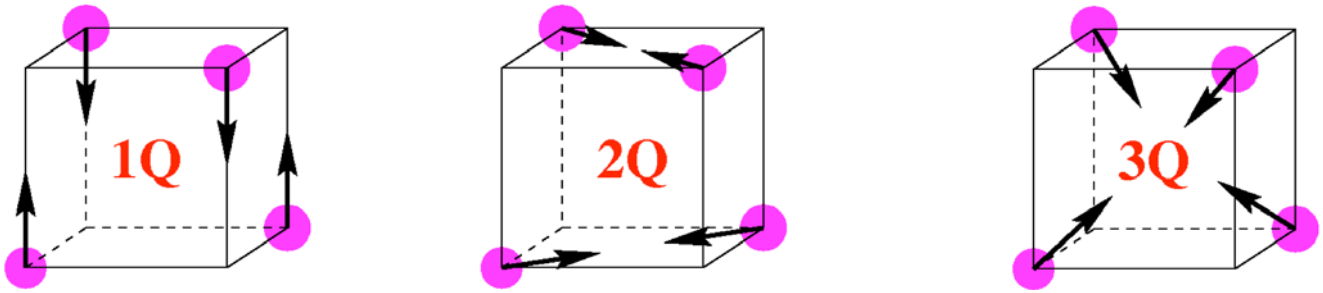
- **FeMn runs on IBM SP3 at NERSC**



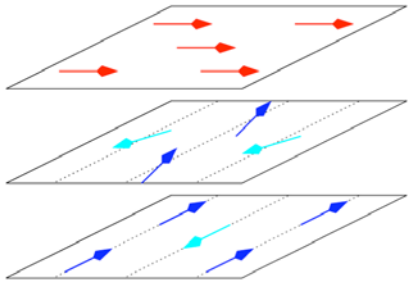
- **Calculations with ~10^4 atoms are possible**
  - **$5nm^3$ has ~ 12,000 atoms**

Magnetism in γ FeMn & FeMn/Py

- **Experimental Summary**
  - **Inelastic Neutron Scattering: FeMn is antiferromagnetically ordered in 1Q, 2Q, and 3Q (Umebayashi and Ishikawa (1966))**
  - **Mossbauer spectroscopy suggests 3Q or 2Q (Kennedy and Hicks (1987))**



  - **But near neighbor Heisenberg model cannot distinguish between these three structures**
- **Perpendicular coupling between FeMn and Permalloy observed (Jungblut et. al. 1995)**
  - **understood in Heisenberg model (Hinchey & Mills 1986, Koon 1997)**



Ab initio calculations for bulk FeMn

- **In a nutshell: 1Q is unstable, 2Q &3Q are stable**

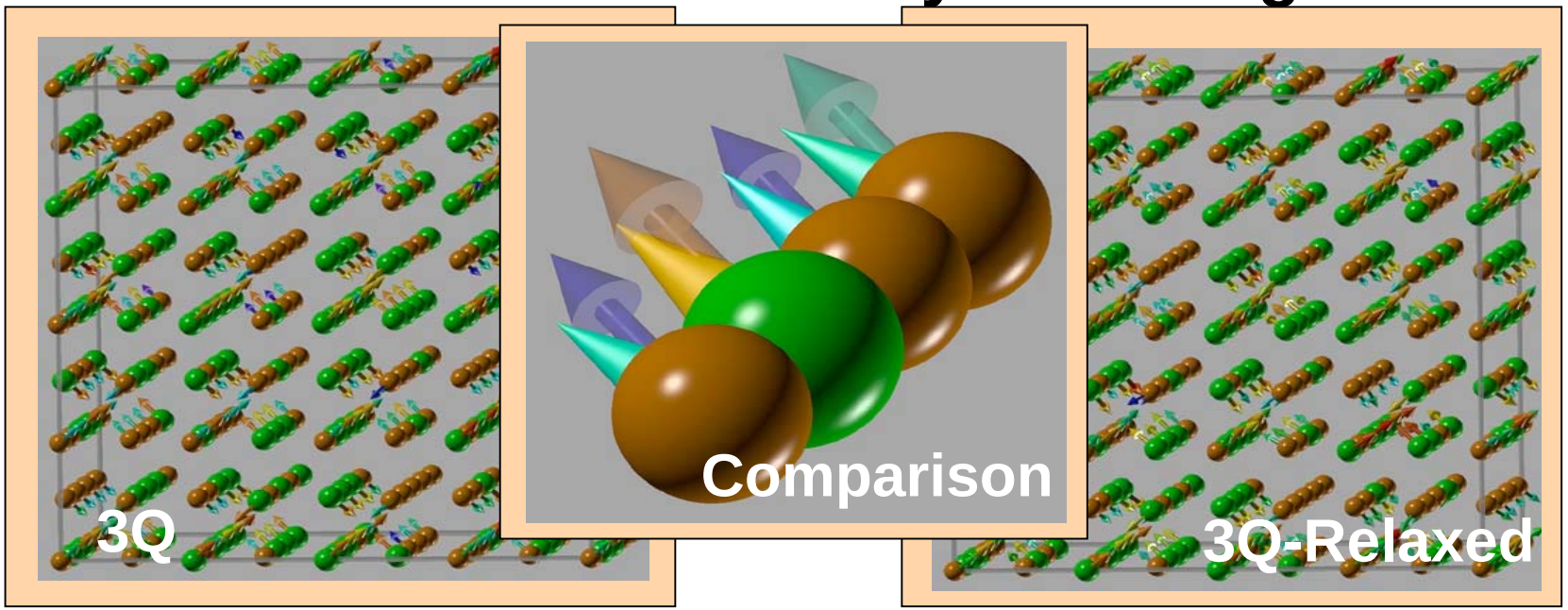
Mean Field (CPA)

KKR-CPA	1 Q	2Q	3Q
μ_{Mn}	1.88	2.00	2.05
μ_{Fe}	1.72	1.85	1.91
E_B(meV)	15.86	5.27	0.0

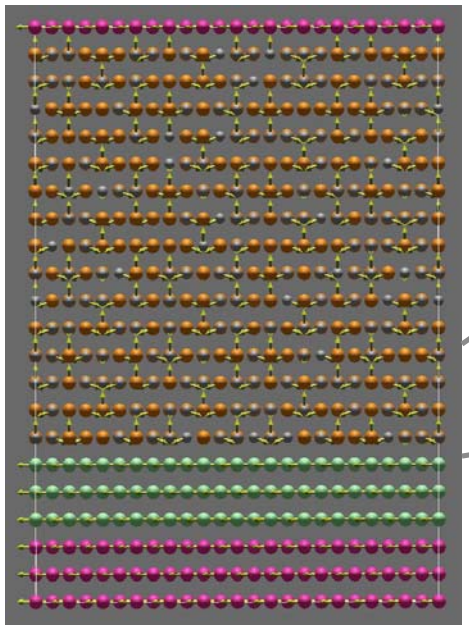
Real Space (super cell)

LSMS	1 Q	2Q	3Q	3Q_R
μ_{Mn}	2.09	2.14	2.17	2.21
μ_{Fe}	1.61	1.75	1.79	1.77
E_B(meV)	15.0	4.14	0.0	-2.5

- **Moment direction varies only a few degrees**



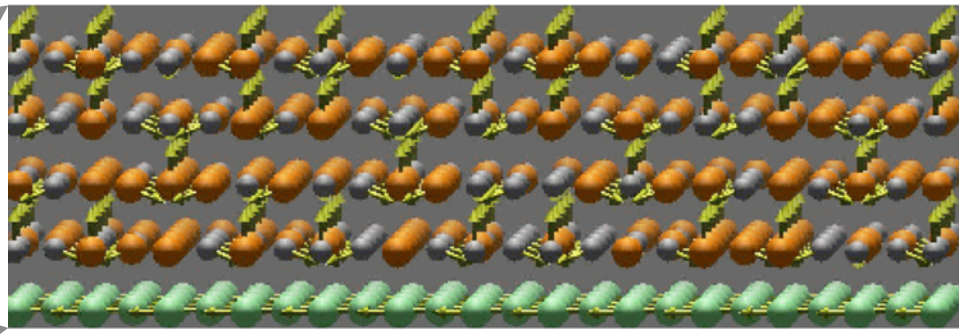
FeMn | Co Multilayers



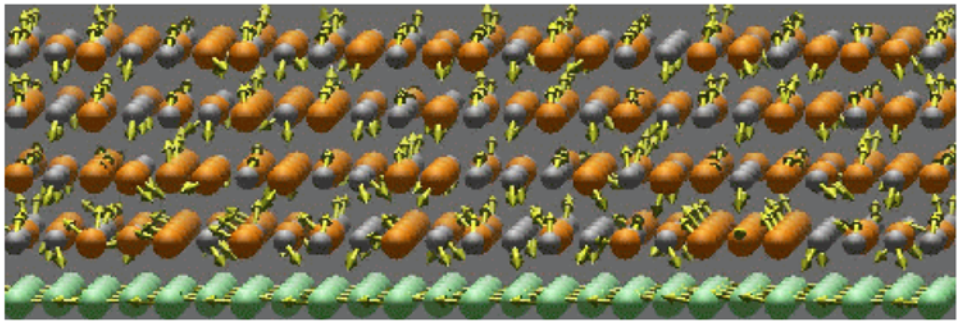
FeMn

Co

Initial state: Ferro for Co, 3Q for FeMn



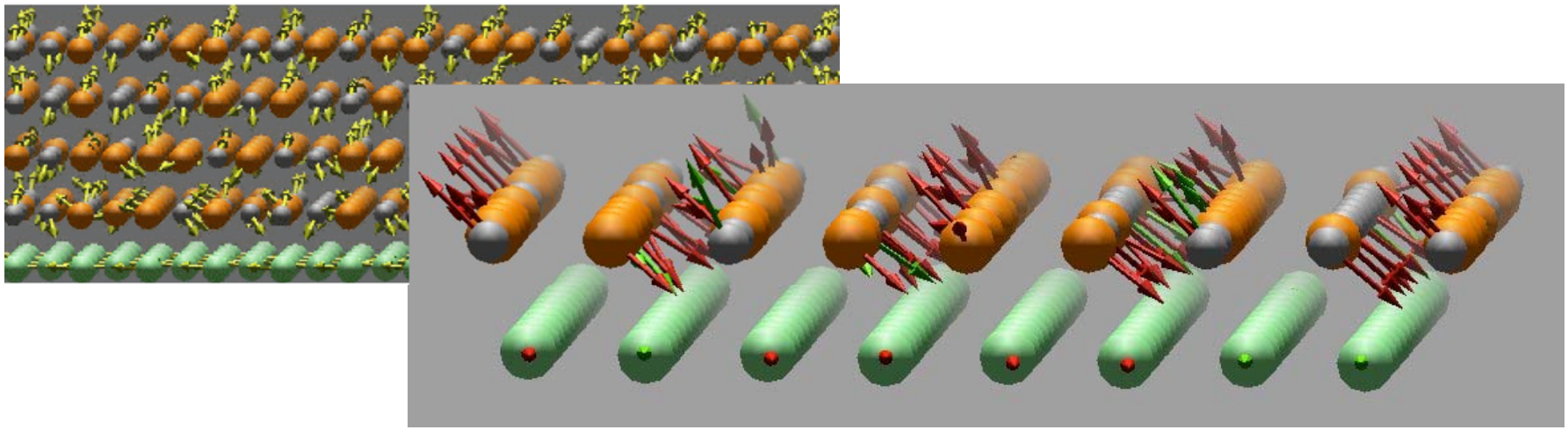
Final State



- **Model of FeMn/Co interface**
  - **15 layers FeMn, random alloy AFM, 3Q**
  - **6 layers Co – 3-frozen FM**
  - **Base 2D cell 12x8**
  - **2016-atom calculation**

Co → **Remains ferromagnetic**
FeMn → **3Q structure destroyed**

Rearranged spin structure due to proximity of FeMn to Co

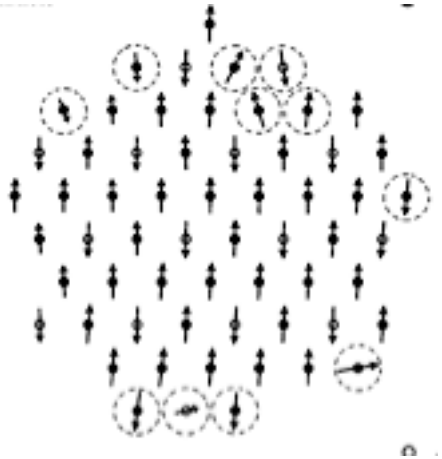


- **Proximity to Co:**
  - **spin structure in FeMn changes from 3Q to 1Q**
  - **FeMn moments are approximately collinear**
- **FeMn moments are perpendicular to Co**
  - **Similar to spin-flop coupling in Heisenberg model**

Looking into the Future

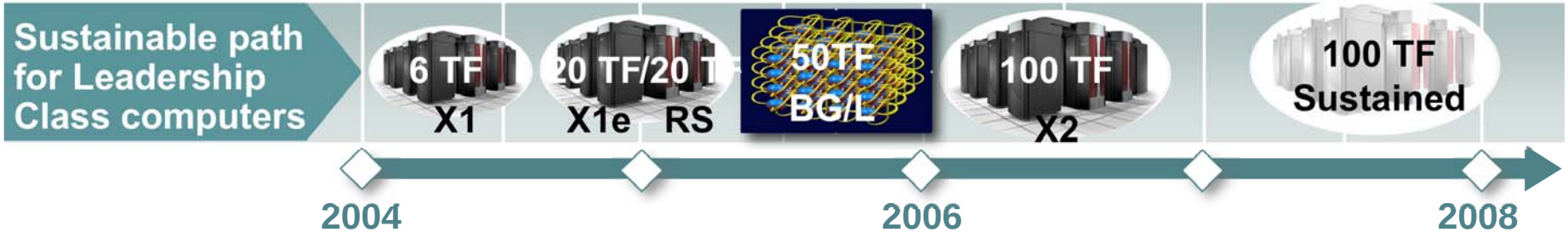
- **In the next 3-5 years supercomputers will be 100-1000 times faster**
  - **Is it really reasonable to scale *ab initio* computation to 10^6 atoms?**
- **Nanomagnets require:**
  - **Ab initio calculations**
  - **Fluctuations at T>0 - entropy effects**
- **Need to calculate free energy**

$$F\left(\vec{e},T\right) = E - T\ln W$$



- **Run multiple *ab initio* calculations in parallel to sample *W***
  - **Probably doable for 100 times faster machine (ongoing project on IBM BG/L)**
- **Examples:**
  - **Temperature dependent effective anisotropy, magnetization, etc. in nanoparticles**
  - **Effective anisotropy for AFM/FM gains (Exchange Bias)**

National Leadership Computing Facility



Instrumentation for Nanoscience

Materials Science Virtual User Center

Materials : Math : Computer Scientists

- *Open Source Repository*
- *Object Oriented Tool Kit*
- *User Laboratories*
- *User training*

User Community

Computational End-Station for Nanomagnetism?

Summary and Conclusions

- **Reliable *ab initio* computation for nano-structures complement experimental probes**
  - **Surface regions dominate nanostructures**
  - **Understand spin structure at the atomic scale**
  - **Materials specific studies**
- **Applying *ab initio* computation to nanostructure is possible**
  - **LSDA to DFT reliable for magnetic materials**
  - **Application to ~10^4 atom currently possible**
- **Future possibilities (3-5 years with 100-1000 x faster computers)**
  - **Dynamics and finite temperature phenomena can be studied at atomic scale**

Acknowledgment

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