

Studying Nanomagnetism with Computational Techniques

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Computational Materials Sciences

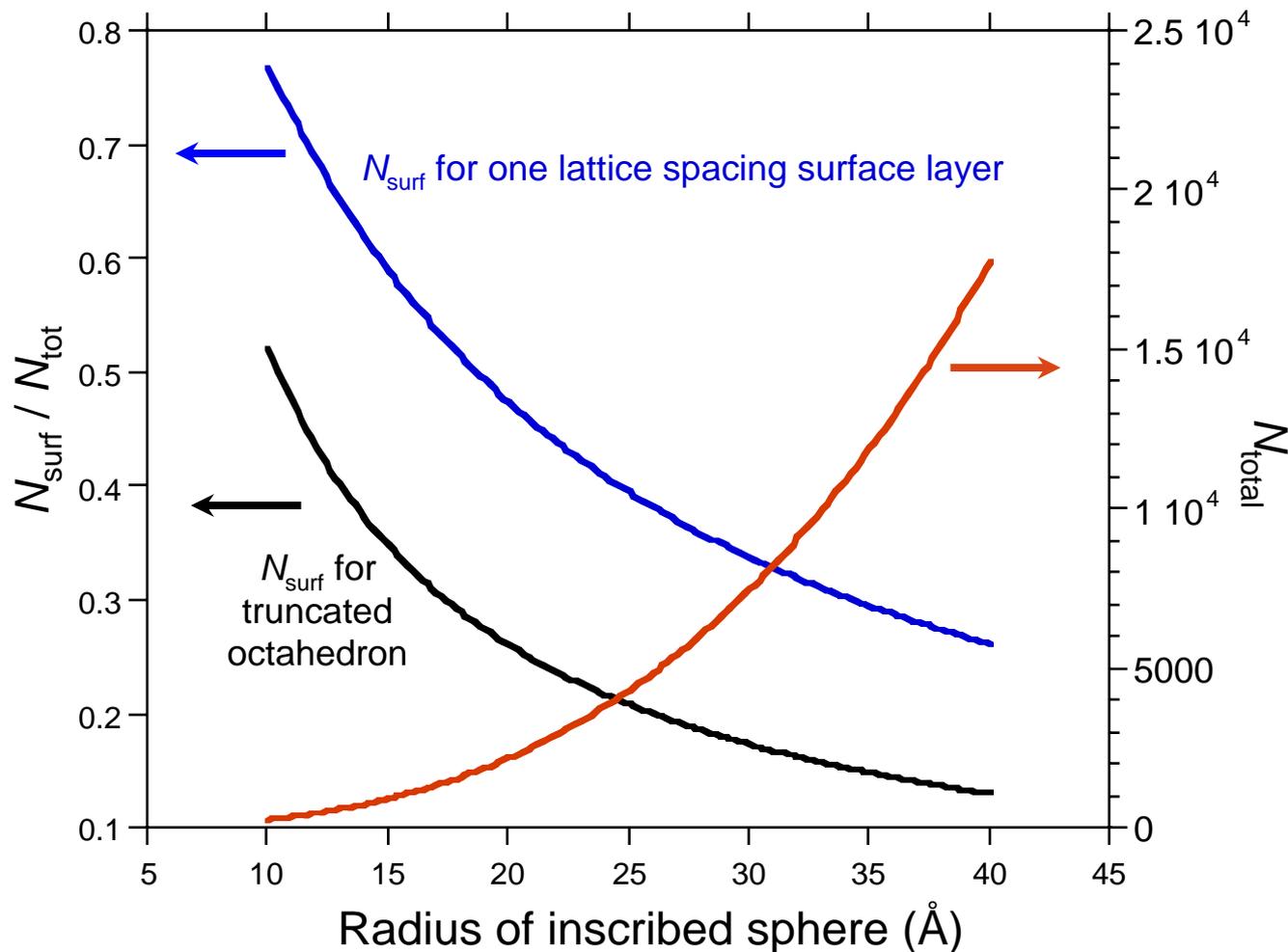
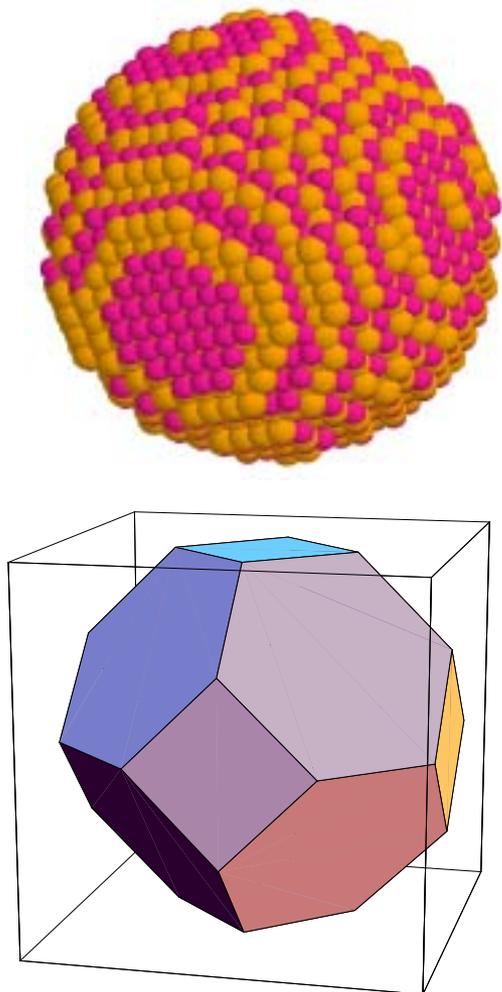
Computer Science and Mathematics Division

Workshop on Nanomagnetism using X-Ray Techniques
Lake Geneva, August 30, 2004

Computational Techniques

- **Transport in magnetic nanostructures**
 - Giant magnetoresistance (Co/Cu/Co)
 - Spin-dependent tunneling (Fe/MgO/Fe)
- **Average magnetic properties**
 - $M_r(T)$ for assemblies of nanoparticles
 - Micromagnetics of patterned nanostructures
- **Statistical physics in nanomagnets**
 - Phase transitions in coupled FM/AFM systems
 - Finite size effects ...
- **Atomic scale magnetic structure / dynamics in nanostructures**

Example: FePt (L10) Nanoparticle



→ Nanoscale is dominated by interface

Surface Spin Disorder in Nanoparticles

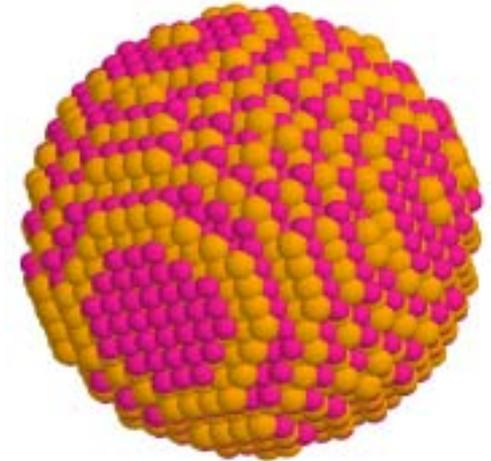
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- **Competition between surface anisotropy and exchange**
- **Maybe even random exchange**
- **Ferro- (Ferri-) magnetic interior couples to spin-glass (antiferromagnetic) surface region**
- **Enhancement of “effective” anisotropy**
- **Note: exchange bias was first observed in oxidized Co particles**

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 nanoparticles!
- 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; ...



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TIFF (LZW) decompressor
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**Complex spin structure of nanoparticles /
nanostructures may be an opportunity**

Atomic scale spin-structure

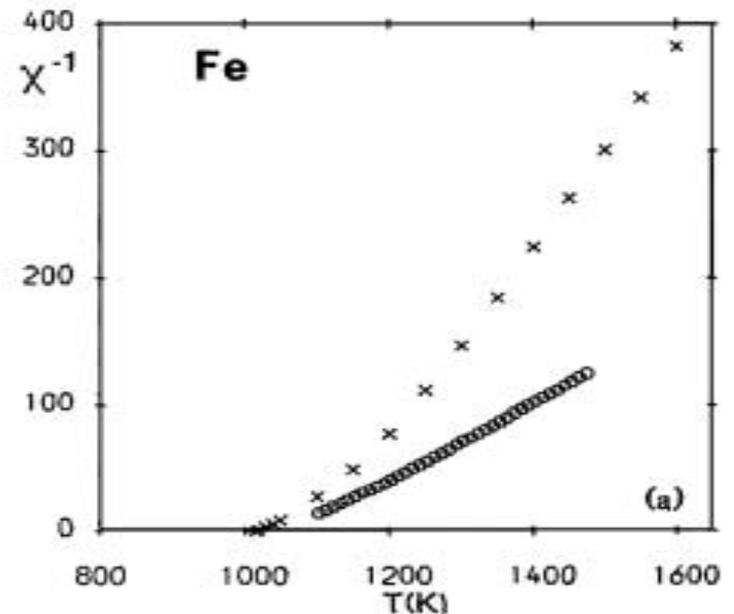
- **Nanomagnetism, opportunities & challenges**
 - need to understand atomic scale spin structure
 - **Experimental probes**
 - Most average over 10-100 or more nanometers
 - Atomic scale spin-structure accessible by inference using models
 - **Models of magnetism at the nano scale**
 - Surface / interface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

In a nutshell, for magnetism: LSDA to DFT is a success!

- **Magnetic moments:**
 - Fe: $\sim 2.3 \mu_B$ exp. $2.2 \mu_B$
 - Co: $\sim 1.6 \mu_B$ exp. $1.7 \mu_B$
 - ... works for many other materials (Ni, Cr, etc.)
- **Similar successes for anisotropy**
- **Curie temperatures:**
 - Mean Field with Onsager Cavity Fields

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

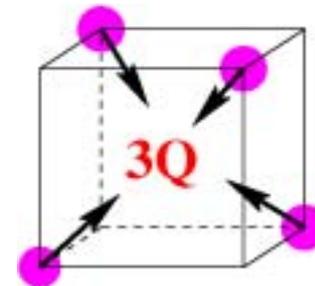
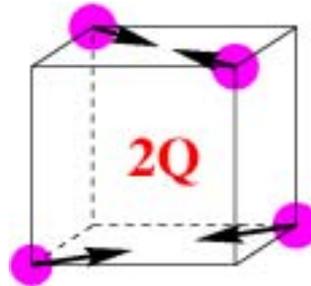
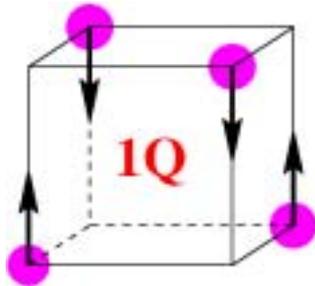
	Fe	Ni
T_c (K)	1015	450
Expt.	1040	631
m_{DLM} (μ_B)	1.89	0.0



First principles calculations: FeMn Bulk

- Experimental Summary

- Neutron Scattering: Antiferromagnetically ordered in 1Q, 2Q, and 3Q (Umebayashi and Ishikawa (1966))
- Mossbauer spectroscopy suggests 3Q or 2Q (Kennedy and Hicks (1987))



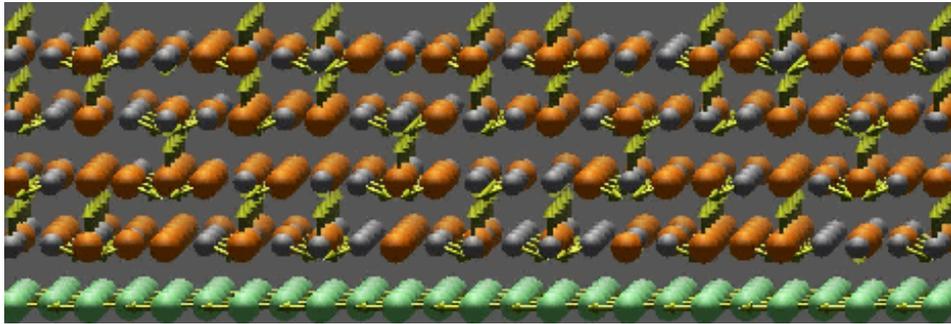
- Results from electronic structure calculations:

- Fe and Mn moments similar
- Predict 3Q is ground state
- 1Q state is unstable

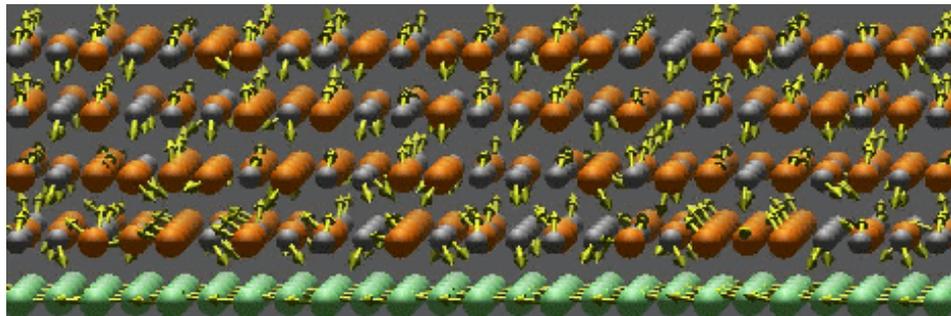
	Mean Field (CPA)			Real Space (LSMS)			
	1 Q	2Q	3Q	1Q	2Q	3Q	3QR
μ_{Mn}	1.88	2.00	2.05	2.09	2.14	2.17	2.21
μ_{Fe}	1.72	1.85	1.91	1.61	1.75	1.79	1.77
E(meV)	15.86	5.27	0.0	15.0	4.14	0.0	-2.5

First principles calculations: FeMn/Co bilayers

Initial state: Ferro for Co, 3Q for FeMn

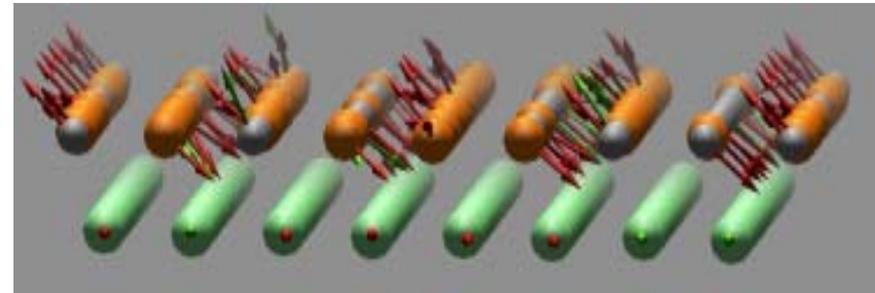


Final (relaxed) State



Co → Remains ferromagnetic
FeMn → 3Q structure destroyed

- Proximity to Co changes spin magnetic structure in FeMn from 3Q to 1Q
- Fe and Mn moments are approximately collinear (1Q)
- Co moments are perpendicular to Fe and Mn moments
 - Same as spinflop coupling



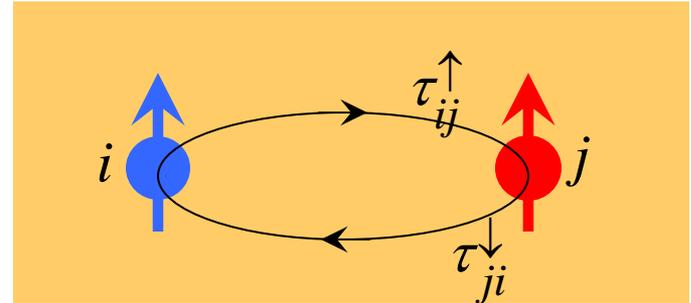
Calculate Exchange Interaction

Lichtenstein et. al. JMMM 1987

Heisenberg exchange: $E_{ij} = -J_{ij} \hat{s}_i \cdot \hat{s}_j$

Embed to moment into FM (AFM) host

Calculate change in energy upon small
Perturbation of moment direction:



$$J_{ij} = -\frac{1}{4\pi} \text{Im} \int_{-\infty}^{E_F} \text{Tr}_L \left(\Delta_i \tau_{ij}^{\uparrow} \Delta_j \tau_{ji}^{\downarrow} \right) d\varepsilon$$

Sum rule:

$$J_i = \sum_{j \neq i} J_{ij} \text{sgn} \left(\hat{s}_i \cdot \hat{s}_j \right) = -\frac{1}{4\pi} \text{Im} \int_{-\infty}^{E_F} \text{Tr}_L \left(\Delta_i \left[\tau_{ii}^{\uparrow} - \tau_{ii}^{\downarrow} \right] + \Delta_i \tau_{ii}^{\uparrow} \Delta_i \tau_{ji}^{\downarrow} \right) d\varepsilon$$

Exchange interactions for Fe, Co, and Ni

Mean field estimate for $T_c = \frac{2}{3} \frac{1}{k_B} J_i$ $J_i = \sum_j J_{ij}$

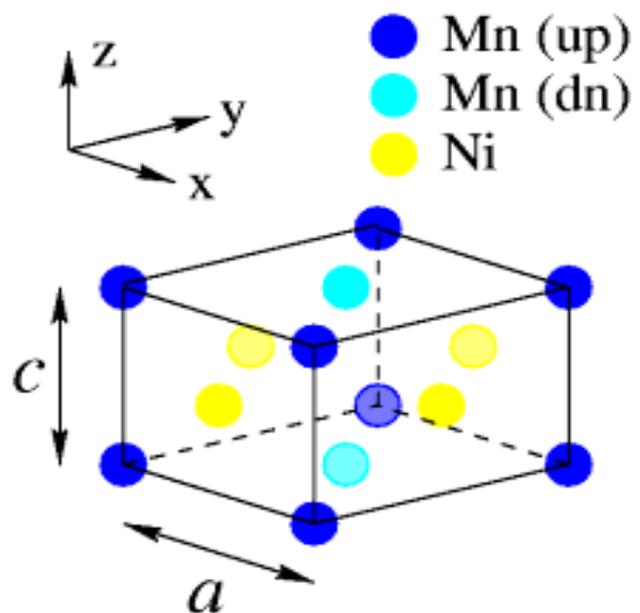
	J_i (meV)	T_c	T_c (experiment)
Co	179.6	1375 K	1388 K
Fe	169.6	1312 K	1043 K
Ni	41.6	322 K	631 K

LSDA results and exchange in NiMn

Use lattice parameters for thin films:

$$a=3.697\text{\AA} \quad c/a=0.9573$$

Magnetic moments: $\mu_{\text{Ni}}=0$ $\mu_{\text{Mn}}=3.2\mu_{\text{B}}$ (experiment: $3.8\pm 0.3\mu_{\text{B}}$)



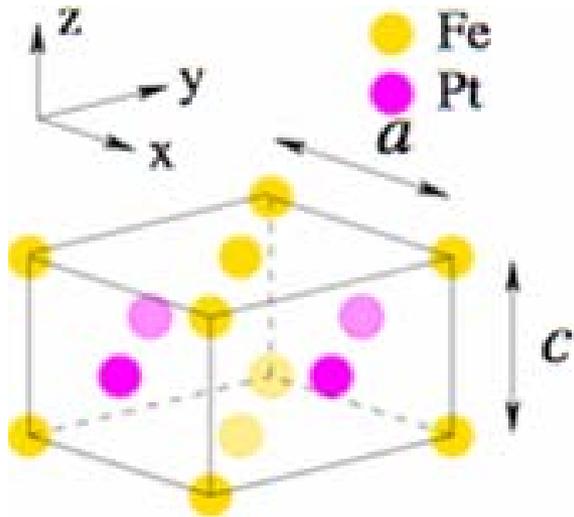
Mean field $T_{\text{N}}=1187\text{K}$

(experiment: $1070\pm 40\text{K}$)

R_{ij}	$J_{ij}(\text{meV})$	$J_i(\text{sum})$
(.5,.5,0.)	-27.1	109.3
(0.,0.,1.)	16.1	141.8
(1.,0.,0.)	6.7	168.4
(.5,.5,1.)	1.8	152.1
(0.,1.,1.)	-2.3	134.4
(1.,1.,1.)	4.7	153.0

Compare with $J_i(\text{direct}) = 153.4 \text{ meV}$

Exchange Interactions for FePt



$R_{ij}; i=Fe$	$J_{ij} (meV)$	shell
$(.5,.5,0.)$	10.5	42.1
$(1.5,1.5,0.)$	3.0	12.0
$(2.,2.,0.)$	-3.2	-12.9
$(1.,0.,0.)$	10.0	40.1
$(2.5,1.5,0.)$	1.2	9.4
$(.5,.0,.5)$	2.1	19.3
$(0.,0.,1.)$	3.4	6.8
$(.5,.5,1.)$	1.9	15.1
$(1.,0.,1.)$	-6.7	-53.9

- Fe Layers are ferromagnetic
- Net Fe-Fe inter layer coupling is antiferromagnetic

Total energy calculations

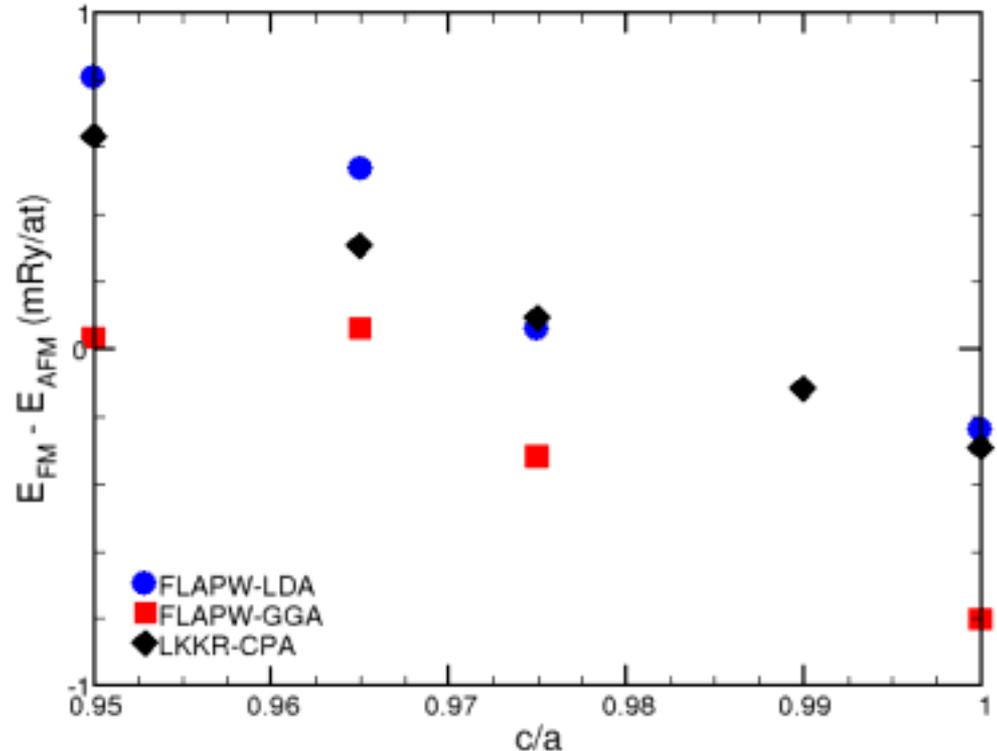
- Total energy calculations give AFM ground state

- KKR: $E_{AF} - E_F = -0.6 \text{ mRy}$

- FLAPW: $E_{AF} - E_F = -0.7 \text{ mRy}$

- Increasing c/a restores FM order

- Generalized Gradient Approximation:
FM and AFM are approx. degenerate



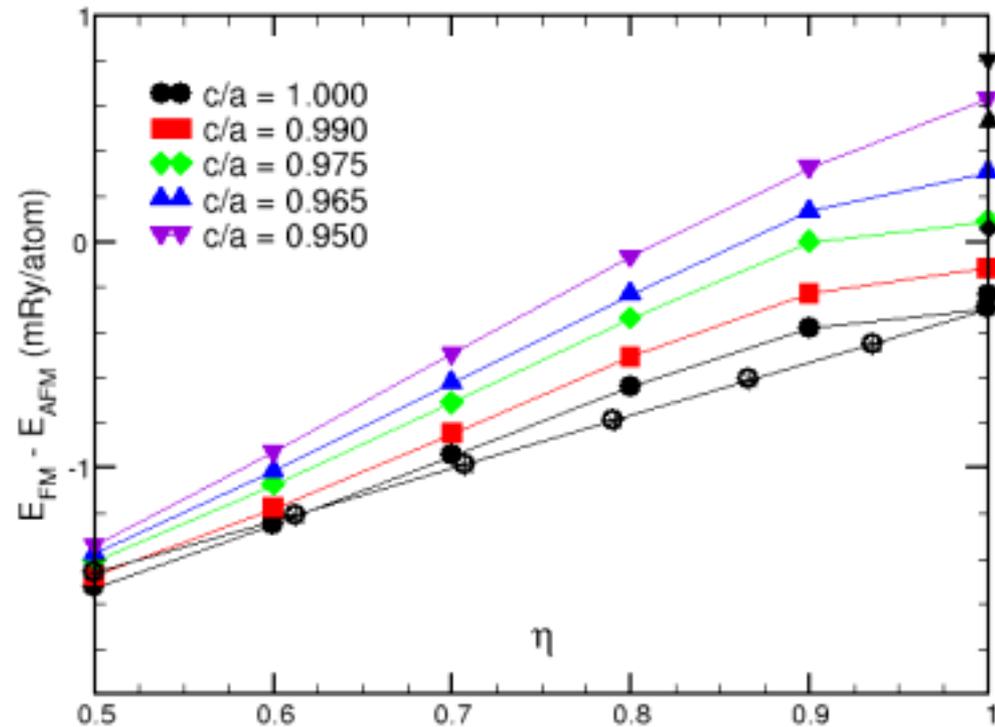
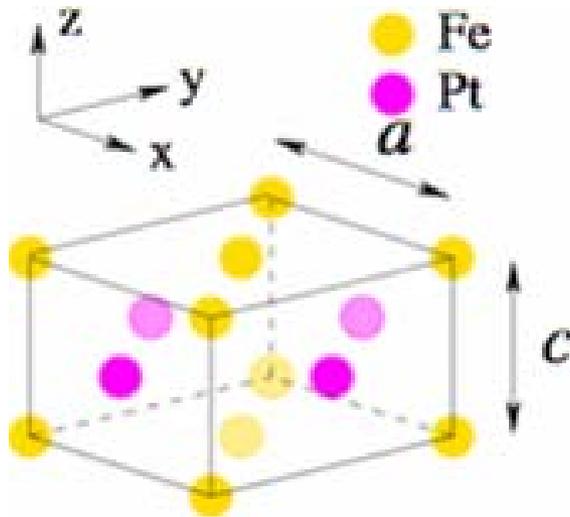
Bulk FePt is close to AFM / FM instability!

Ferromagnetism and anti-site disorder

Long Range Order Parameter:

$$\eta = c[\text{Fe}] - c[\text{Pt}]$$

$$0 \leq \eta \leq 1$$

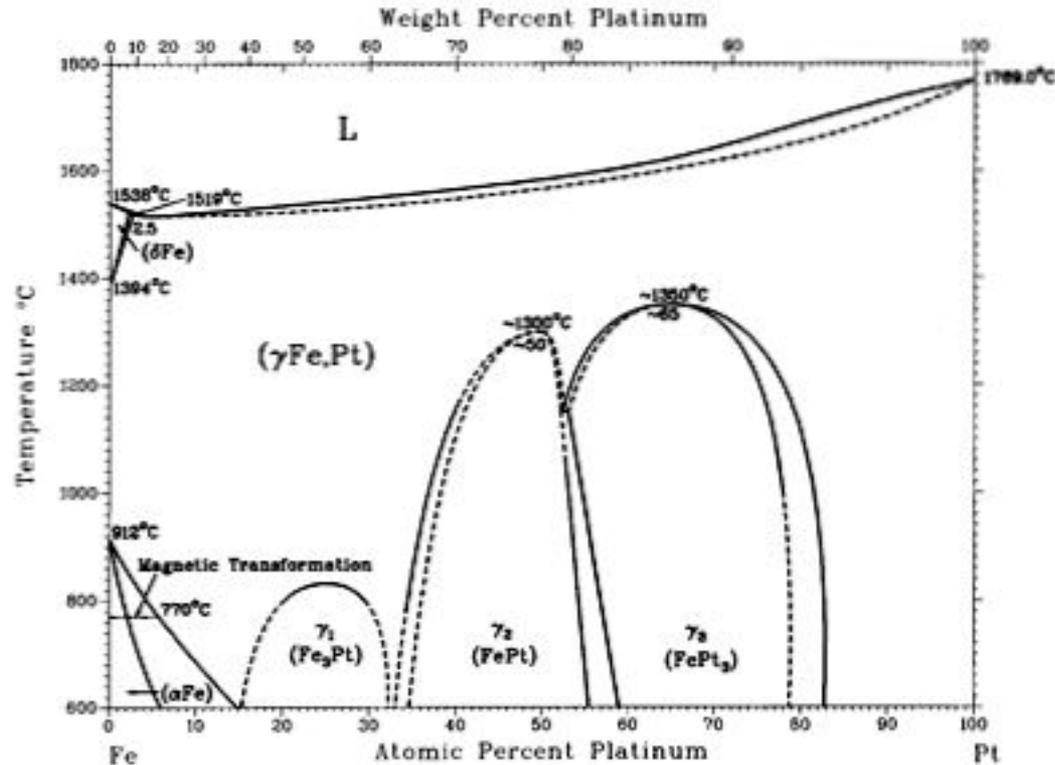


Deviation from perfect order favors ferromagnetism

Fe-Pt Phase Diagram



Fe-Pt Phase Diagram

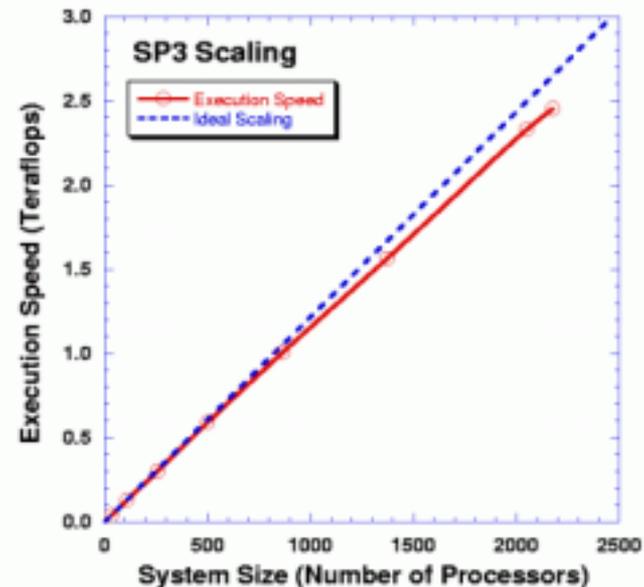


Partial disorder in L10 phase is not unreasonable

High performance computing: Outlook

IBM SP @ NERSC

- Ab-initio spin dynamics (LSMS method)
 - All electron, $O(N)$, scalable
- IBM SP RS6000 (3.26 Tflops)
- Runs (@ 75% sustained efficiency)
 - 2176 processor
 - Magnetic ground state for **~2000 atoms**
 - **2.46 Tflops** (75% efficiency)



Cray X1/X2



- HPC in the near future (3-5 years): (factor of 100 - 1000 faster)
 - IBM BG/L: **10^5 processors**
 - Cray X1/X2: **10^3 faster processors**
- What can we do in the future:
 - Dynamics of single nanoparticles
 - Statistics (free energy, assembly)

What are 10^5 processors good for?

- **Nanomagnets require:**
 - Ab initio calculations
 - Fluctuations at $T > 0$ - entropy effects
- **Need to calculate energy**

$$F(e, T) = E - T \ln W$$

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- **Run multiple ab initio calculations in parallel to sample W**
- **Examples:**
 - Temperature dependent effective anisotropy, magnetization, etc. in nanoparticles
 - Effective anisotropy for AFM/FM gains (Exchange Bias)

Summary and Conclusions

- **Reliable ab initio computation for nanostructures 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 2000-5000 atom currently possible
 - High performance computers a factor 100-1000 faster in the next 3-5 years
- **Dynamics and finite temperature phenomena can be studied at atomic scale**