Temperature dependent free energy barrier for magnetization reversal in nanomagnets

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Collaborators

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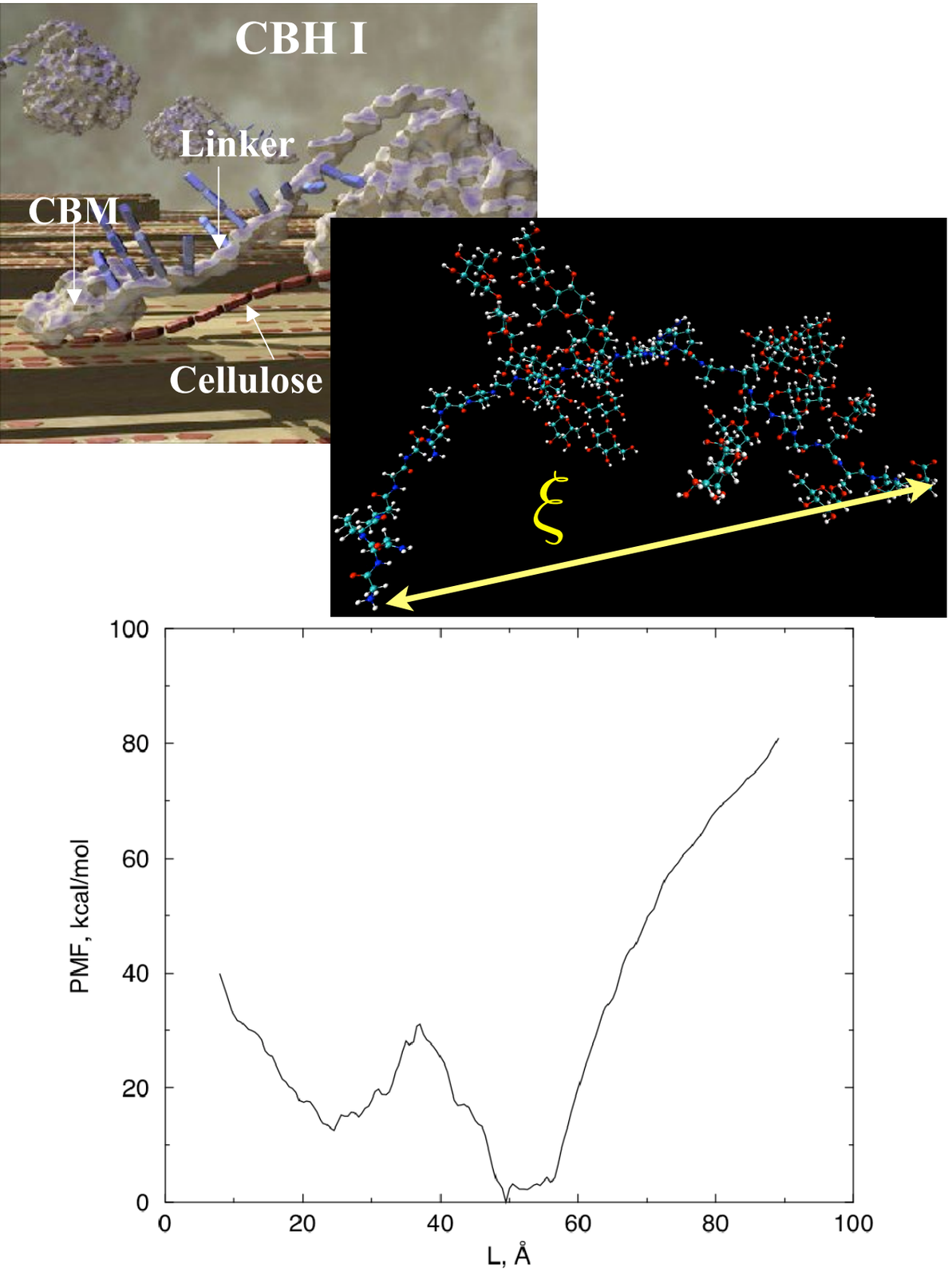
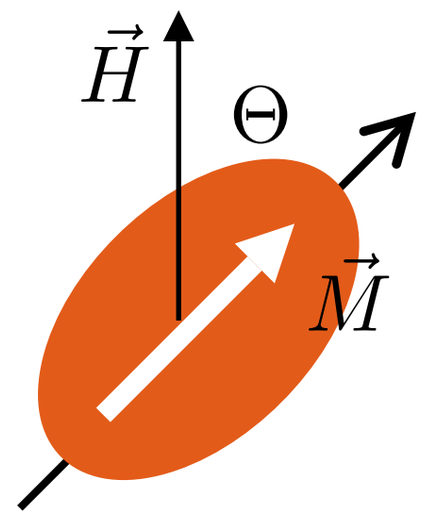
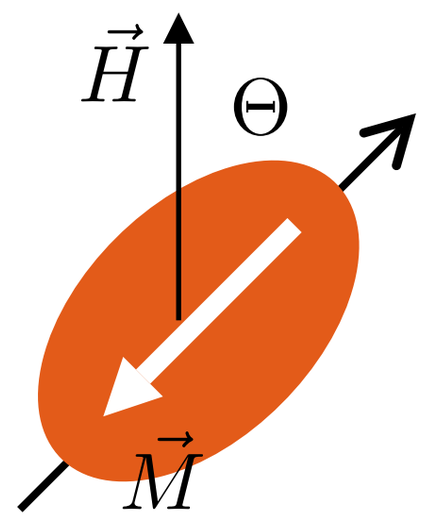
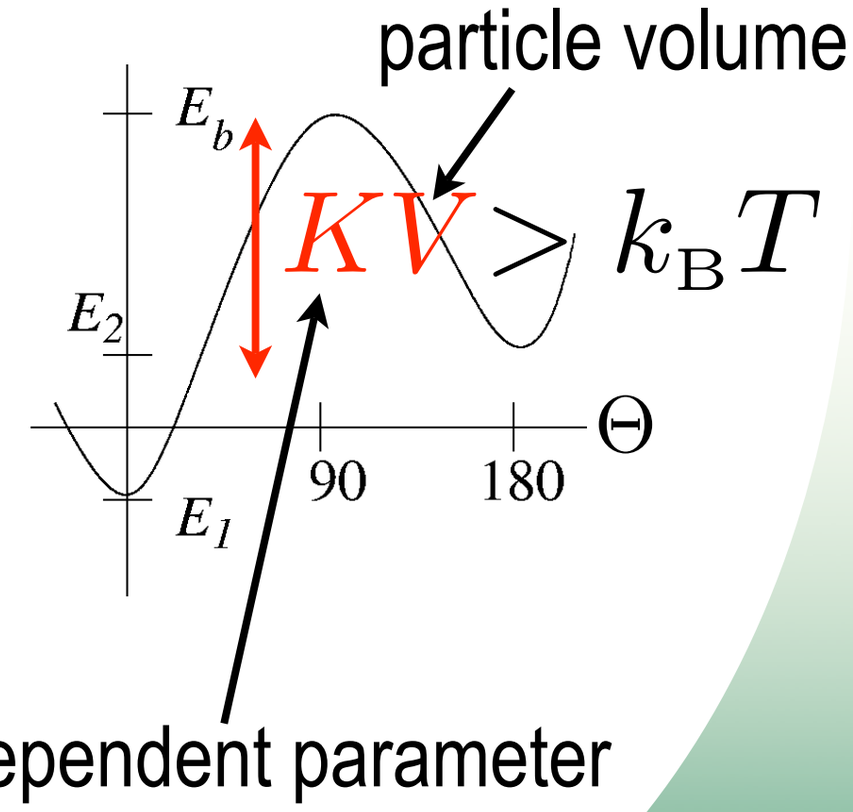
- David Landau, University of Georgia, Athens GA
- Oleg Mryasov, Seagate Research, Pittsburgh PA
- Greg Brown, Tallahassee FL

Free Energy in biophysics and nanomagnetism

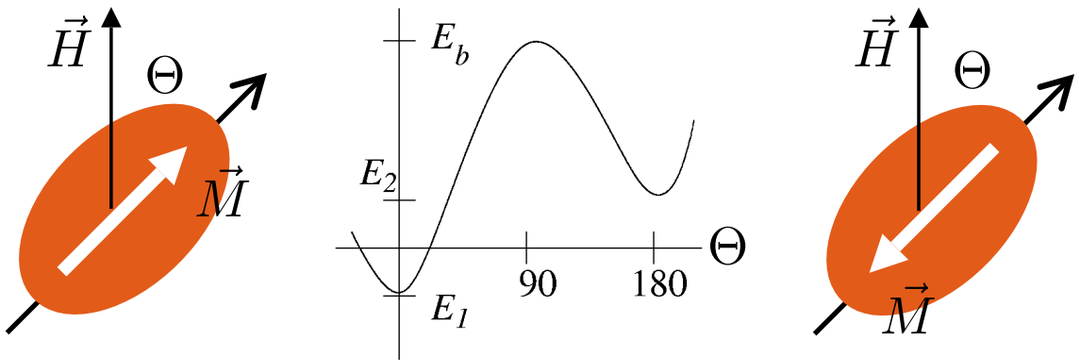


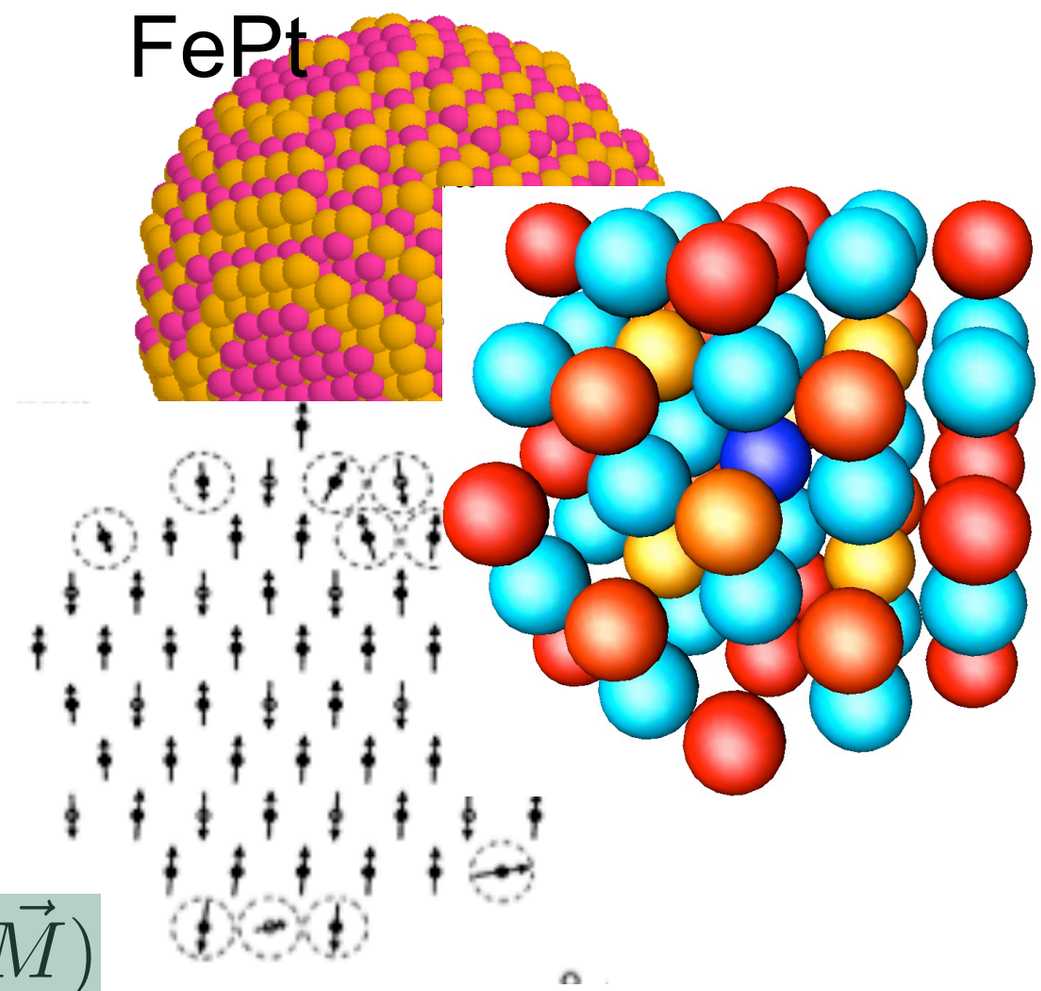






Studying free energy in transition metal magnets





Consider atomic degrees of freedom $\{\vec{m}_1, \vec{m}_2, ..., \vec{m}_N\}$

$$\vec{M} = 1/N \sum_{i=1}^{N} \vec{m}_i$$

$$F(T, \vec{M}) = E(T, \vec{M}) - k_{\mathrm{B}} T \ln W(E, \vec{M})$$

Realistic model or LDA/GGA based DFT calculation. In most cases $E(T, \vec{M}) \approx E(T = 0, \vec{M})$

With the density of states we would know the free energy at all temperature

Can we compute the density of states?

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Metropolis Method

Metropolis et al, JCP **21**, 1087 (1953)

$$Z = \int e^{-E[\mathbf{x}]/k_{\mathrm{B}}T} d\mathbf{x}$$

Compute partition function and other averages with configurations that are weighted with a Boltzmann factor

Sample configuration where Boltzmann factor is large.

1. Select configuration

$$E_i = E[\mathbf{x}_i]$$

2. Modify configuration (move)

$$E_f = E[\mathbf{x}_f]$$

3. Accept move with probability

$$A_{i\to f} = \min\{1, e^{\beta(E_i - E_f)}\}$$

Wand-Landau Method

Wang and Landau, PRL **86**, 2050 (2001)

$$Z = \int W(E) e^{-E/k_{\mathrm{B}}T} dE$$

If configurations are accepted with probability $1/W$ all energies are visited equally (flat histogram)

1. Begin with prior estimate, eg $W'(E) = 1$
2. Propose move, accepted with probability

$$A_{i\to f} = \min\{1, W'(E_i)/W'(E_f)\}$$

3. If move accepted increase DOS

$$W'(E_f) \to W'(E_f) \times f \qquad f > 1$$

4. Iterate 2 & 3 until histogram is flat
5. Reduce $f \to f = \sqrt{f}$ and go back to 1

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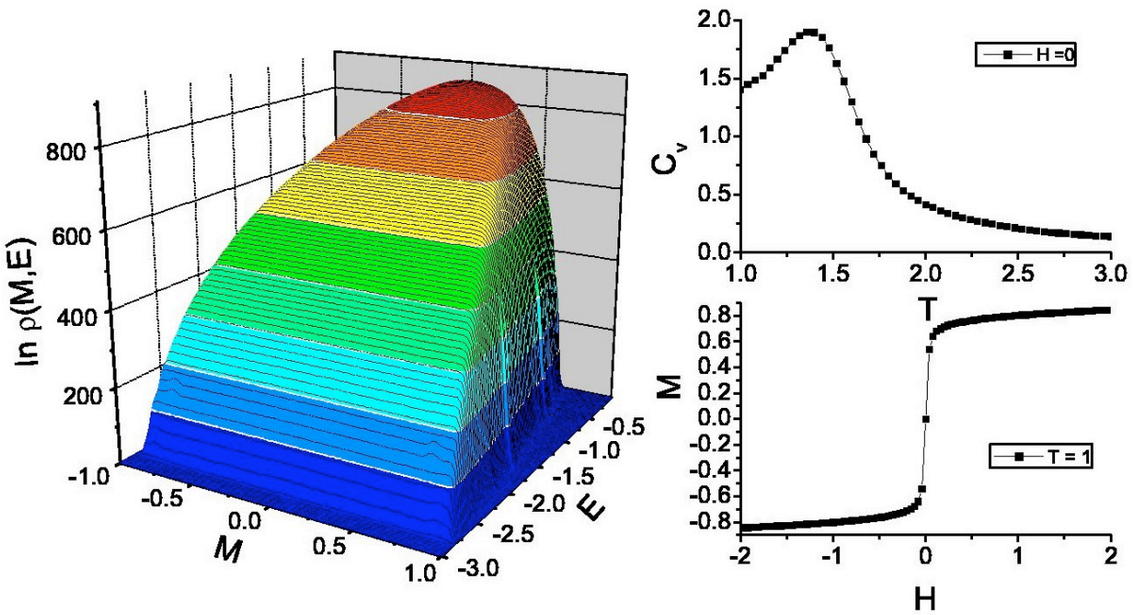
Generalization to continues models with applications to magnetism, proteins, ...

Zhou, Schulthess, Torbrügge, and Landau, Phys. Rev. Lett. **96** 120201 (2006)



Heisenberg model:

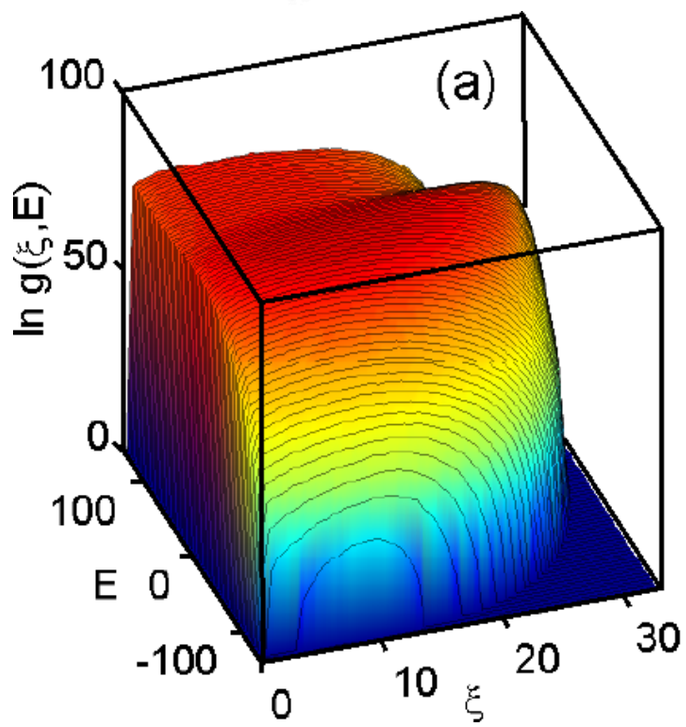
$$W(E, M)$$

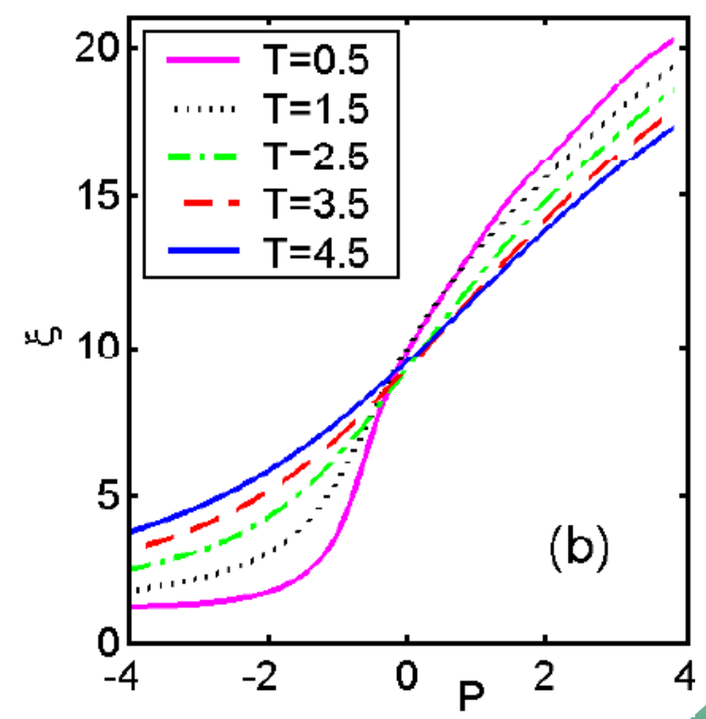
magnetization

Chain with torsional forces

$$W(E, \xi)$$

end-to-end distance





Metropolis Method

Wand-Landau Method

$$Z = \int e^{-E[\mathbf{x}]/k_\mathrm{B}T} d\mathbf{x}$$

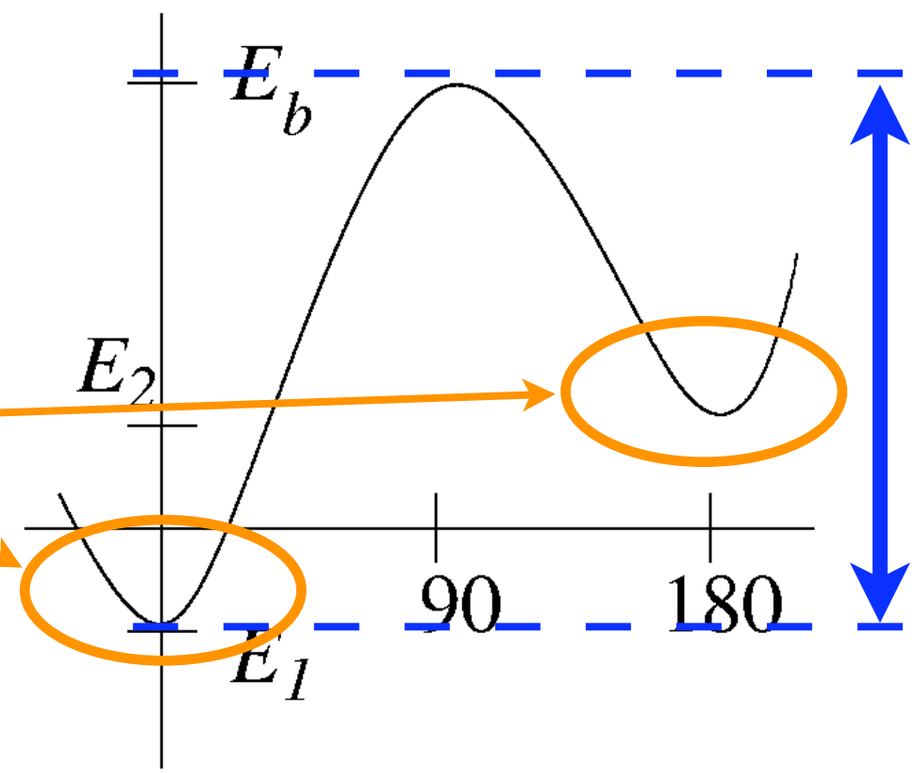
$$Z = \int W(E) e^{-E/k_\mathrm{B}T} dE$$

Sample configuration space with probability

$e^{-E[\mathbf{x}]/k_\mathrm{B}T}$

$1/W(E[\mathbf{x}])$

Samples mainly regions around energy minima



Samples all energies equally

The main features of our model for FePt

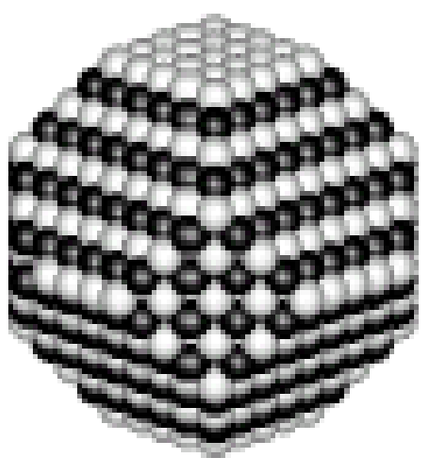
Mryasov, et. al. Europhys. Lett. 86, 805 (2005)

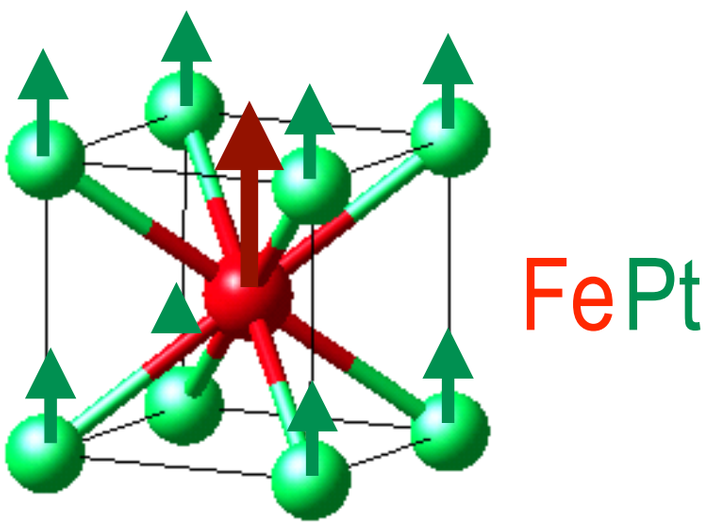
Standard Heisenberg form with uniaxial anisotropy

$$E(\{\vec{m}_i\}) = -\sum_{i<j,\alpha} J_{ij,\alpha} m_{i,\alpha} m_{j,\alpha} - \sum_i K_i m_{i,z}^2$$

Only Fe spins are free variables (induced moments on Pt)

Truncate exchange interaction at the surface





When all spins are aligned, energy has the standard form

$$E = KV sin^2\Theta$$

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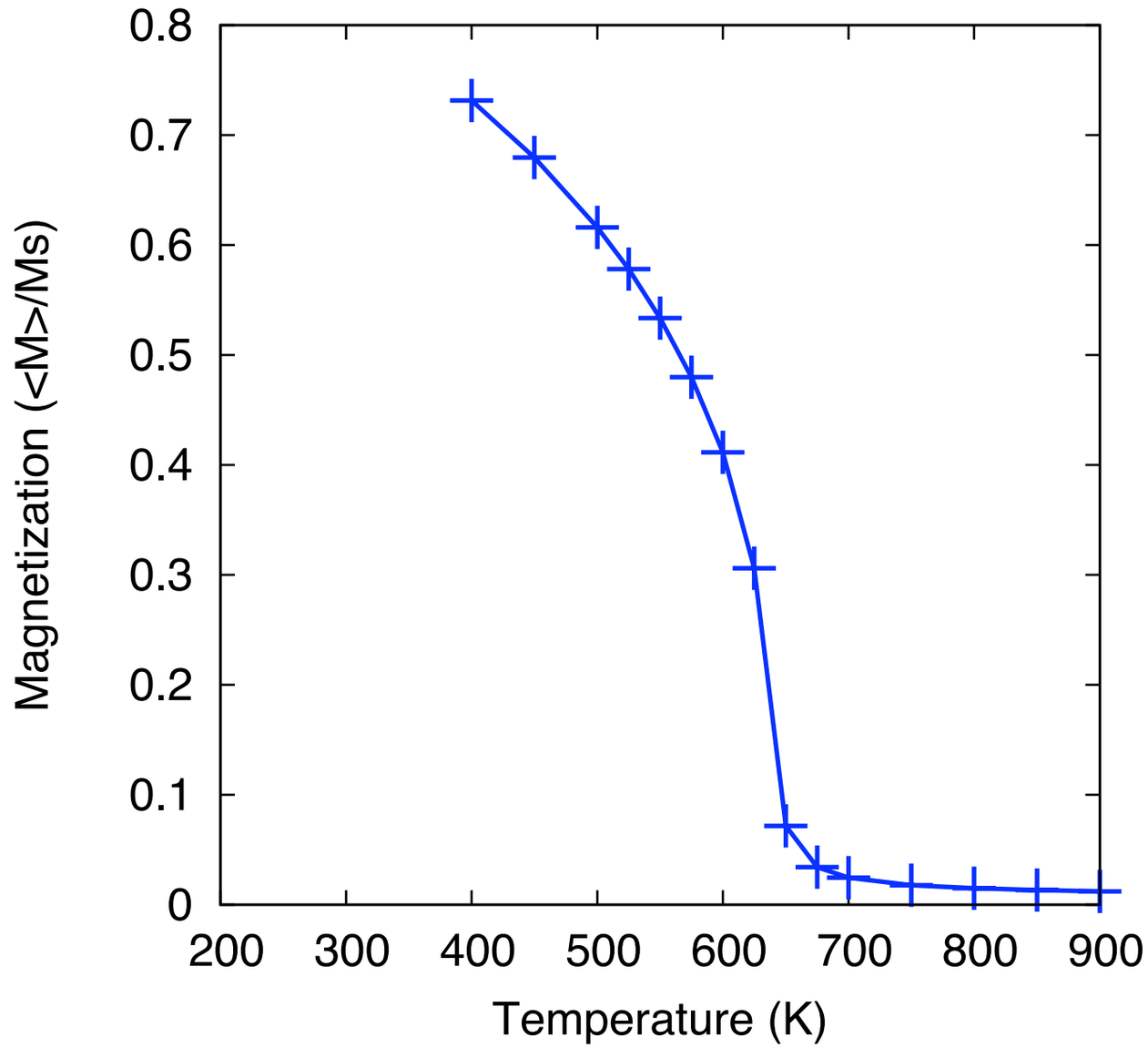
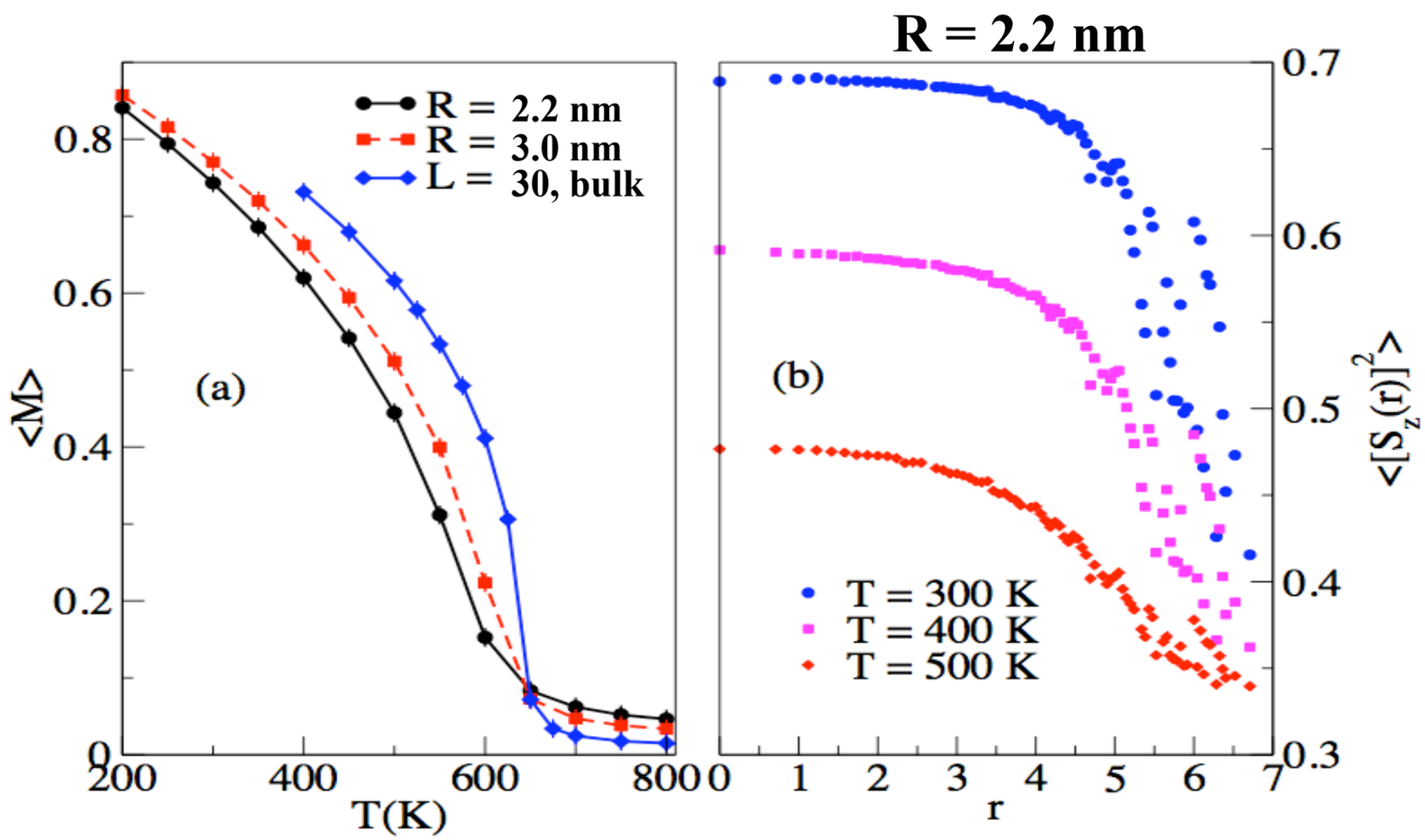
Test the model for bulk FePt (L=30) with standard Metropolis Monte Carlo

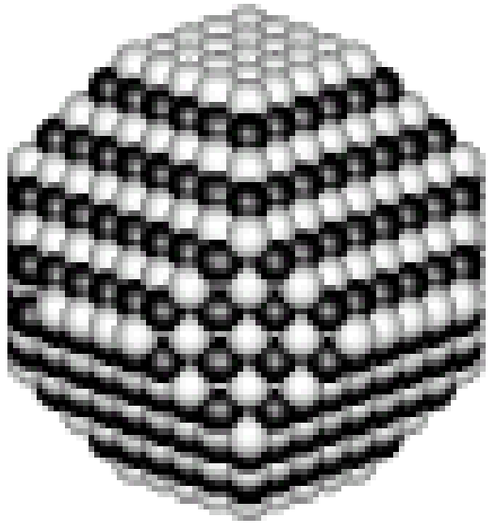


Curie temperature within about 10% of experiment

Ising type phase transition

Temperature dependent magnetization for nanoparticles





- Small effect on Curie temperature
- Magnetization suppressed in surface region
  - Expected due to truncation of exchange interactions at surface
- Surface region does have structure
  - Due to shape of nanoparticle (truncated octahedron)

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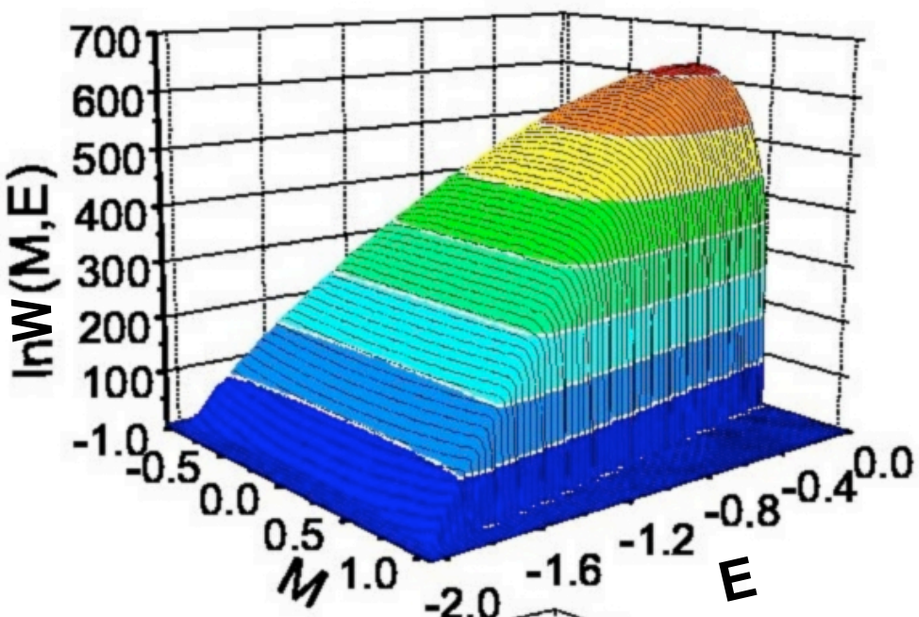
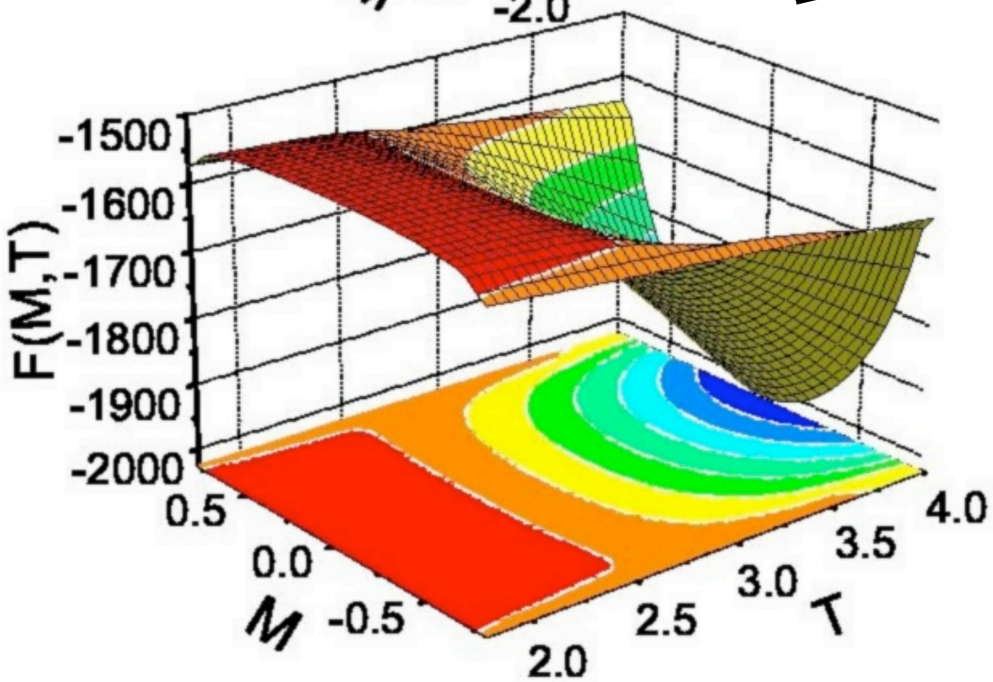
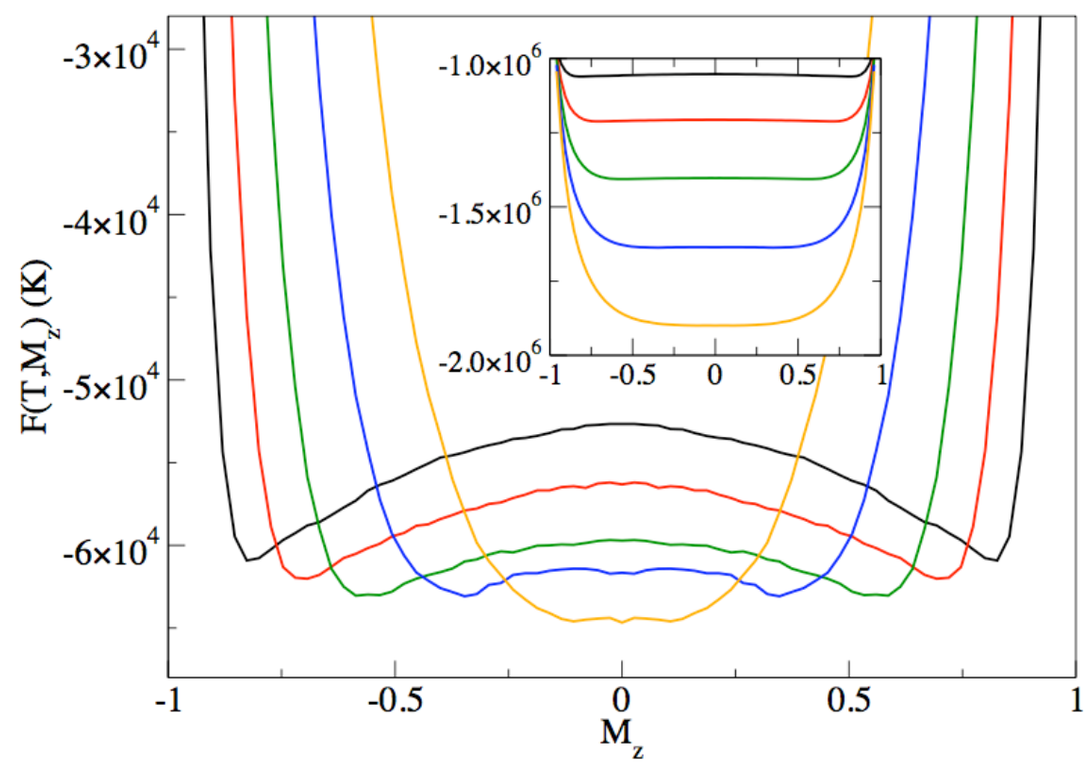
Joint density of states and free energy





Numerically integrate density of states

$$F(M_z, T) = -k_{\mathrm{B}} T \ln \int W(M_z, E) \exp(-E/k_{\mathrm{B}}T) dE$$



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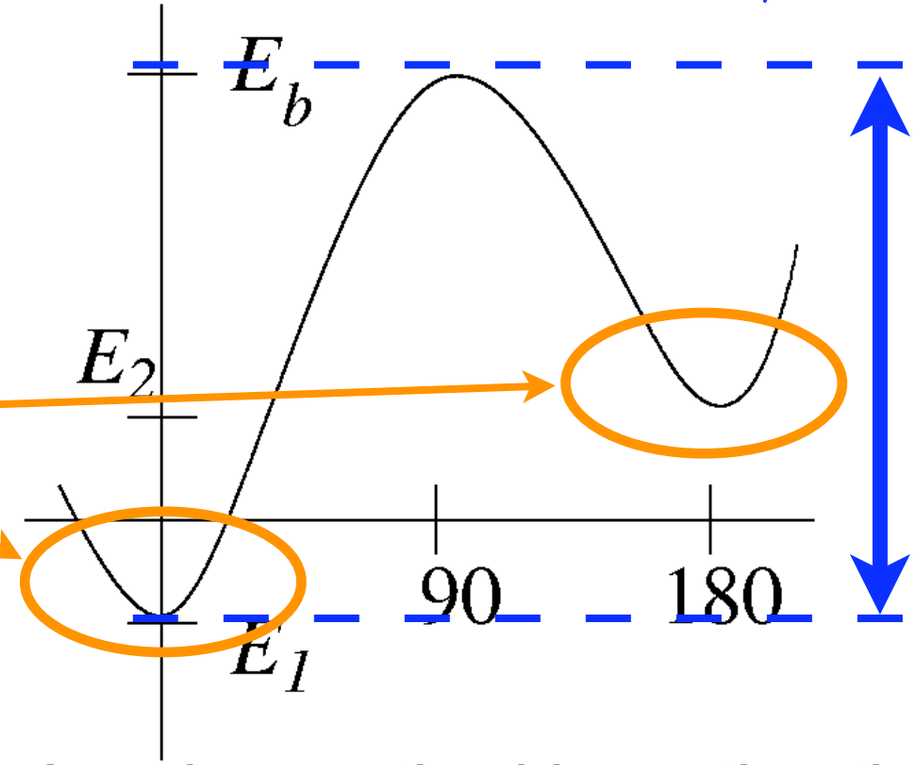
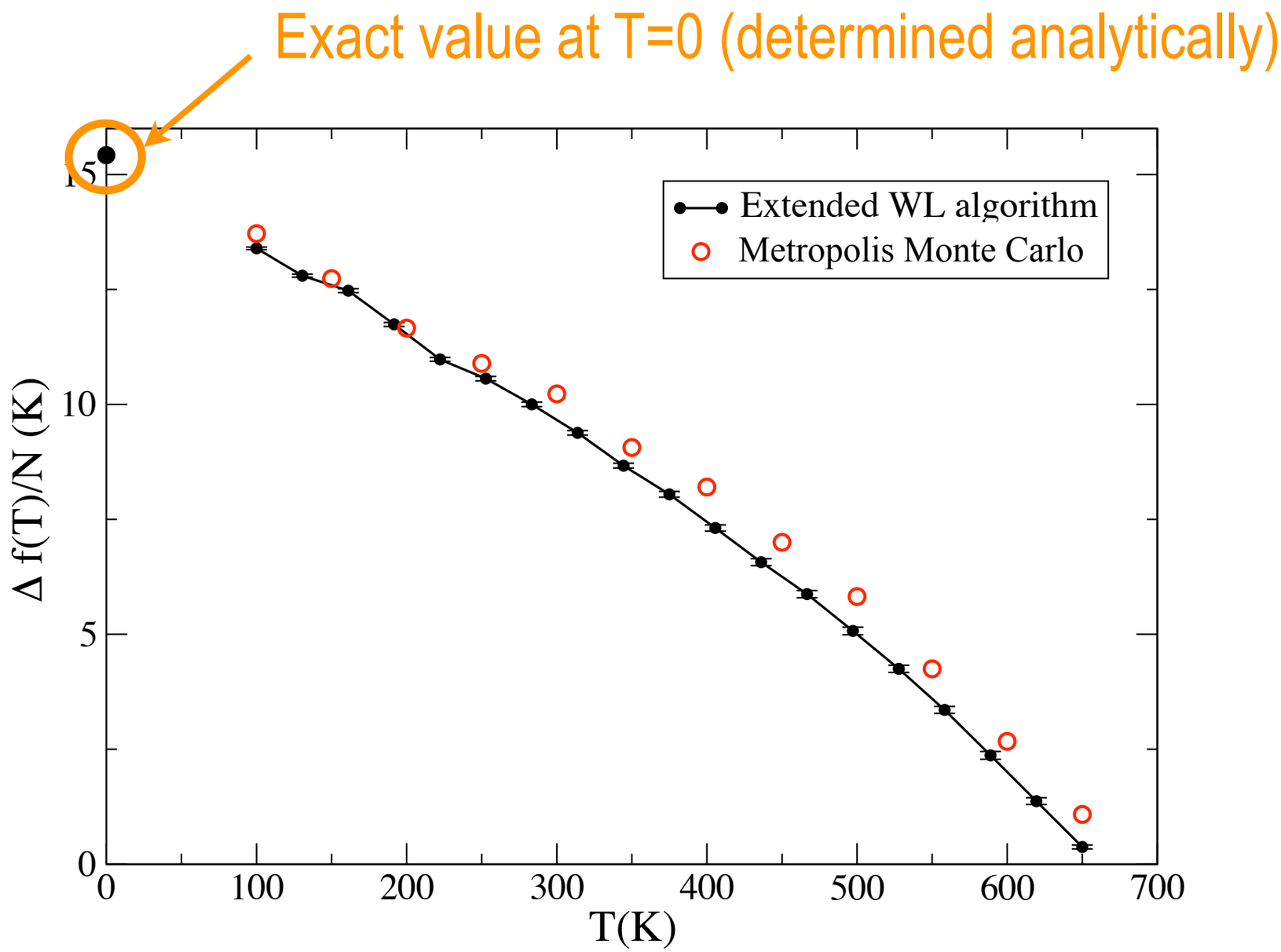


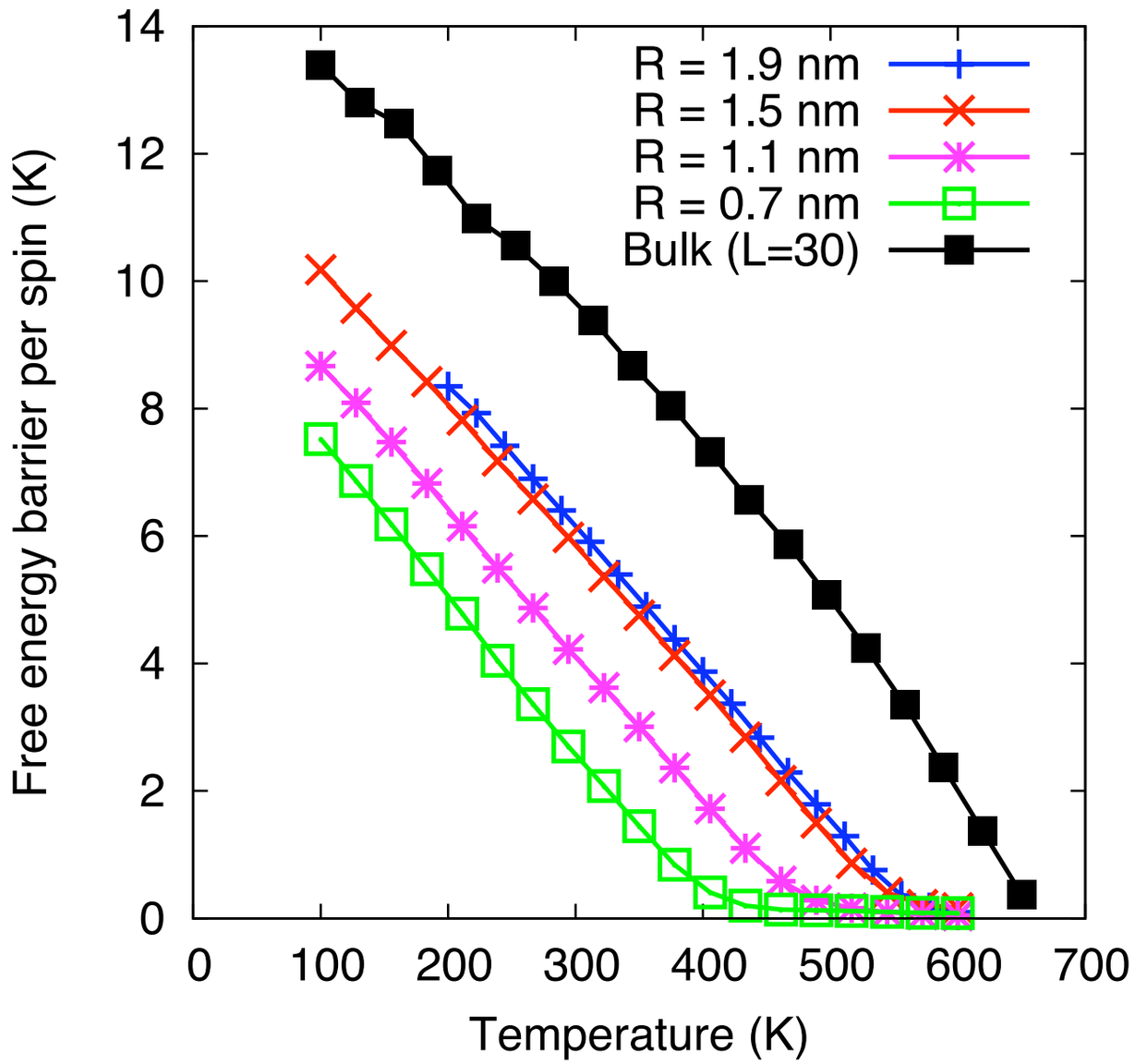
Samples all energies equally -

Check validity of Wang-Landau method by estimating barrier hight from Metropolis MC and fitting to $KV \sin^2 \Theta$

Quantitative test for bulk FePt



Temperature and particle size dependent magnetic free energy barrier



- Quantitative reduction of barrier hight in nanoparticles.
- Linear temperature dependence of barrier in nanoparticles.

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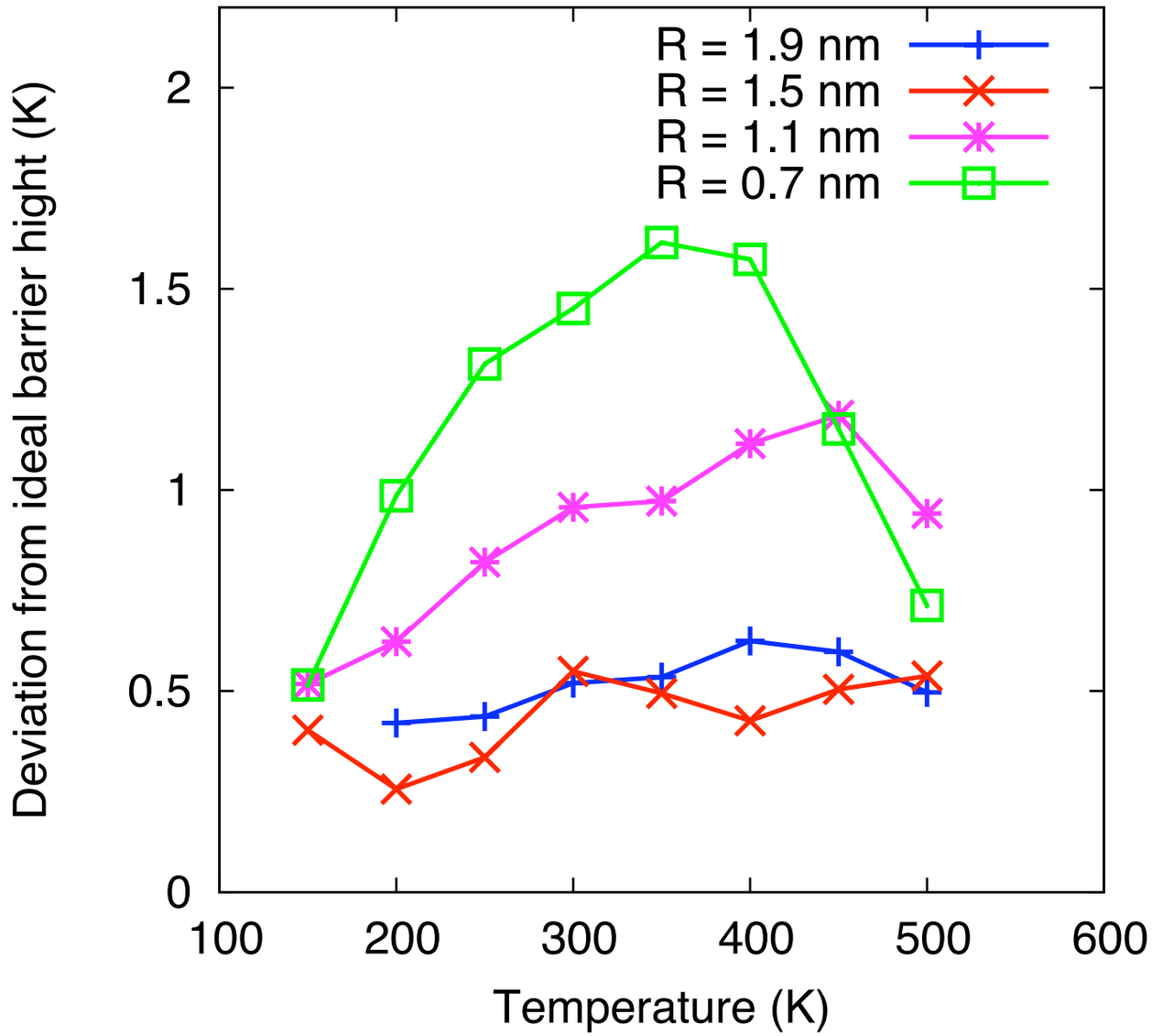
Deviation from idealized Stoner-Wolfarth behavior.

Plot the deviation of the barrier hight from the idealized Stoner-Wolfarth behavior $KV\sin^2\Theta$



Incoherent modes are important in smallest particles - large surface to volume ratio.

Summary and outlook

- Temperature dependent free energy barrier in nanomagnets: immediate relevance to data storage applications
- Direct computation of the joint density of states is possible for systems with ~10^3 microscopic degrees of freedom
- Reliable computation of temperature free energy barrier
- Preliminary model calculations show interesting nano-scale effect that need to be investigated with first principles methods (LDA/GGA)
- Outlook: combine Wang-Landau with ab initio DFT (LSMS)
  - will scale perfectly to 10^6 cores!
  - WL/LSMS code will run at >80% of peak floating point performance

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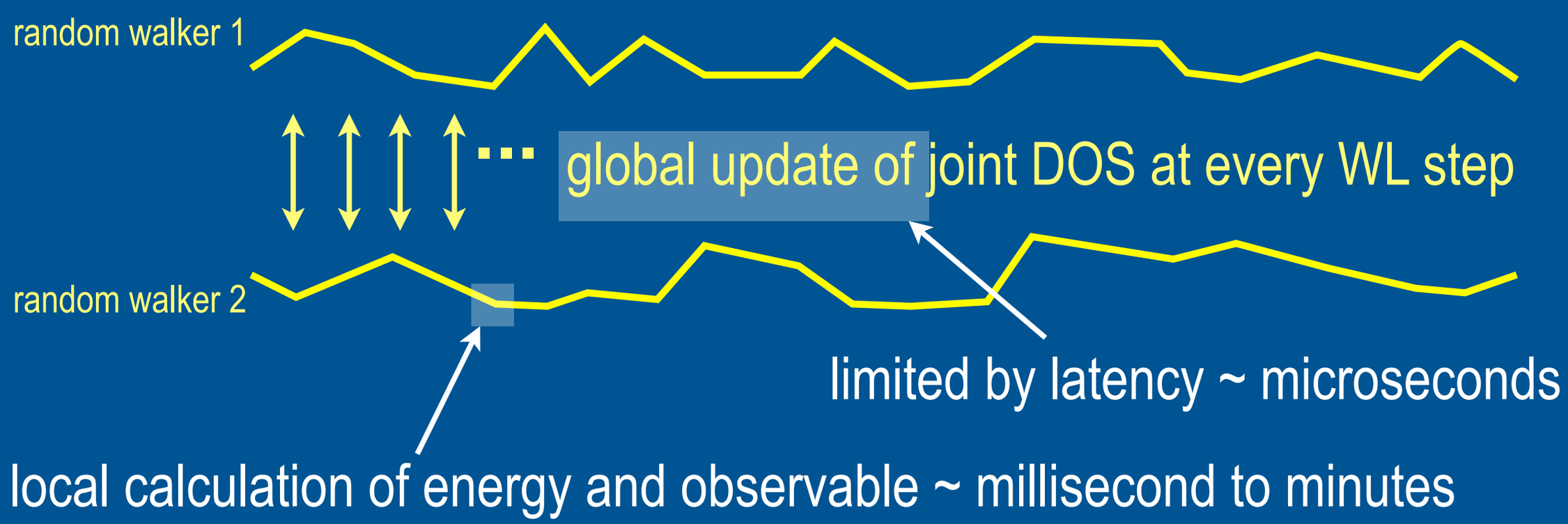
Wang-Landau method is NOT embarrassingly parallel

Metropolis MC acceptance: $A_{i\to f} = \min\{1, e^{\beta(E_i - E_f)}\}$

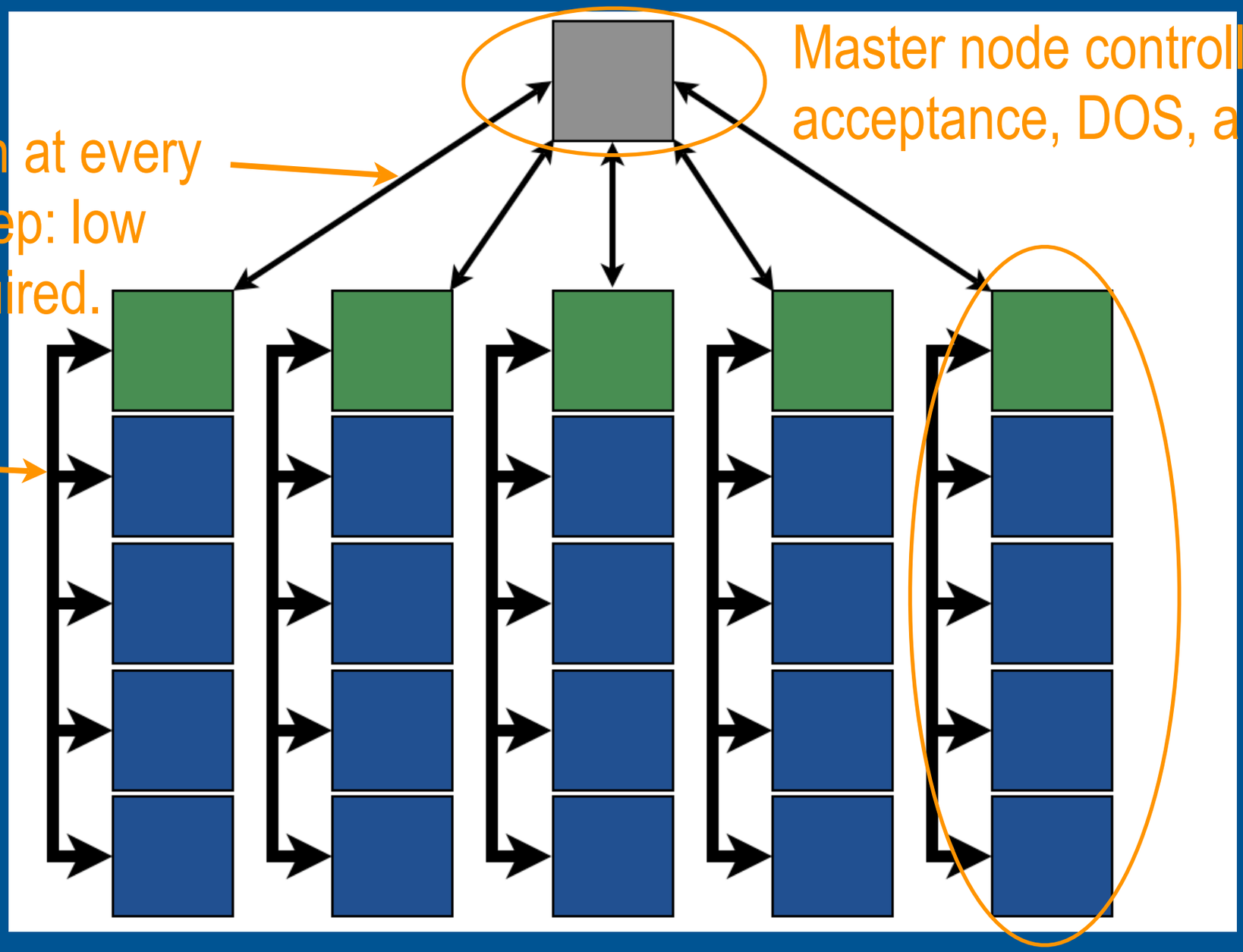
Generalized Wang-Landau acceptance:

$$A_{i\to f} = \min\{1, W'(E_i, X_i)/W'(E_f, X_f)\}$$



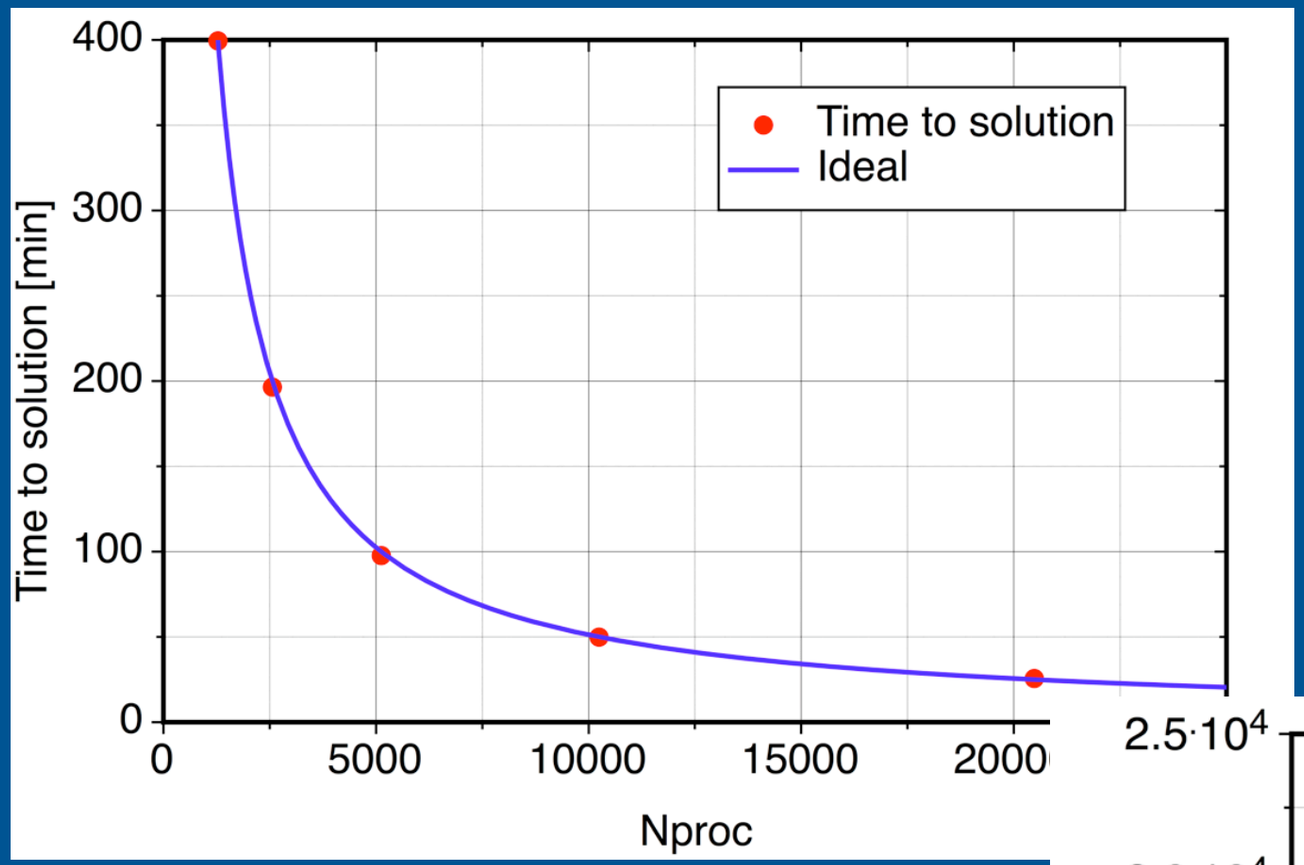
Hybrid communication model

Not embarrassingly parallel!

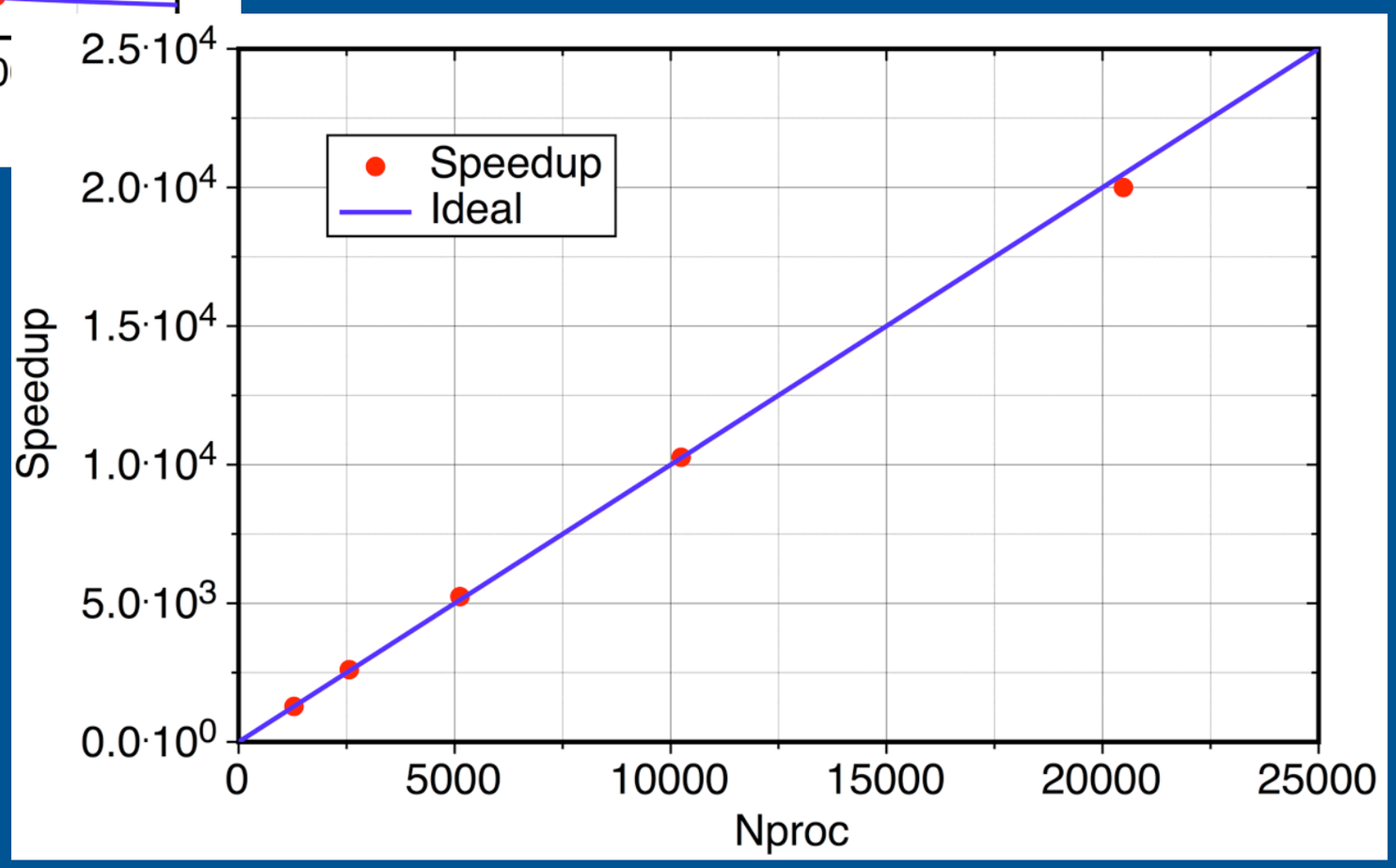


Very general as Wang-Landau method can be used in conjunction with any simulation method (DFT, MD/MM, spin models ...)

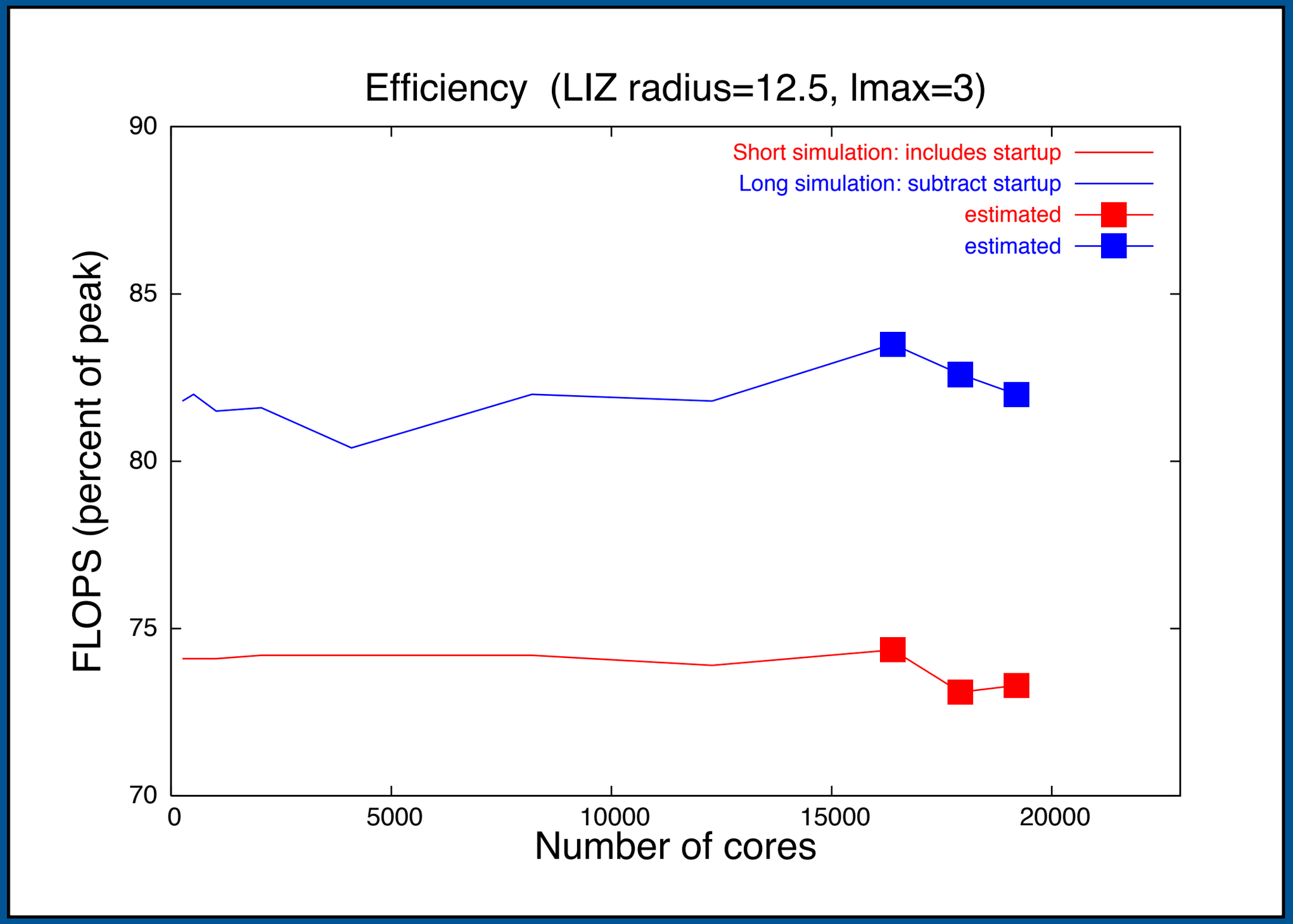
Strong scaling results for hybrid WL-LSMS code



128 Fe atoms and 800 Monte Carlo samples running on Cray XT4 (Jaguar)

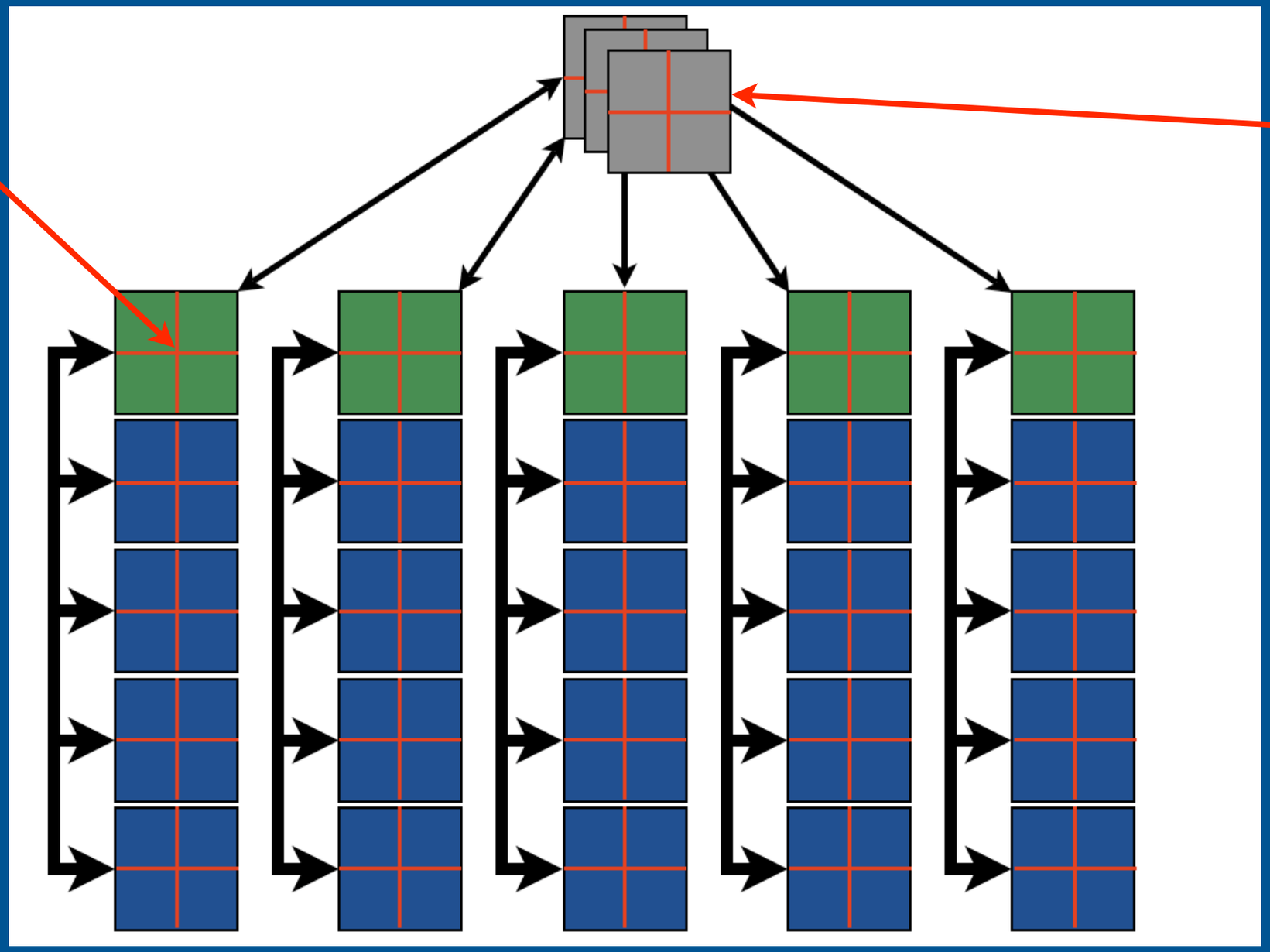


Efficiency of gWL/LSMS prototype code



Extensions to the communication model

1. Take advantage of multi-core nodes



2. Consider and prepare for fault tolerance.

3. Parallelize master - asynchronous WL algorithm?