Temperature-dependence of magnetism of free Fe clusters

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Outline

- Introducing finite-temperature magnetism (a bit of theory)

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- Dependence of mean magnetization of clusters on temperature and on cluster size
- Critical (“Curie”) temperature T_c of clusters
- Temperature-dependence of magnetic profiles of clusters

Motivation

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follow the *coordination number* !

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Lowering the coordination number

$\longrightarrow$	increase of magnetic moments	$\longrightarrow$	enhanced magnetization
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When comparing surfaces with bulk:

competition between enhancement of magnetic moments
and reduction of their coupling

Bulk, surfaces, clusters, . . .

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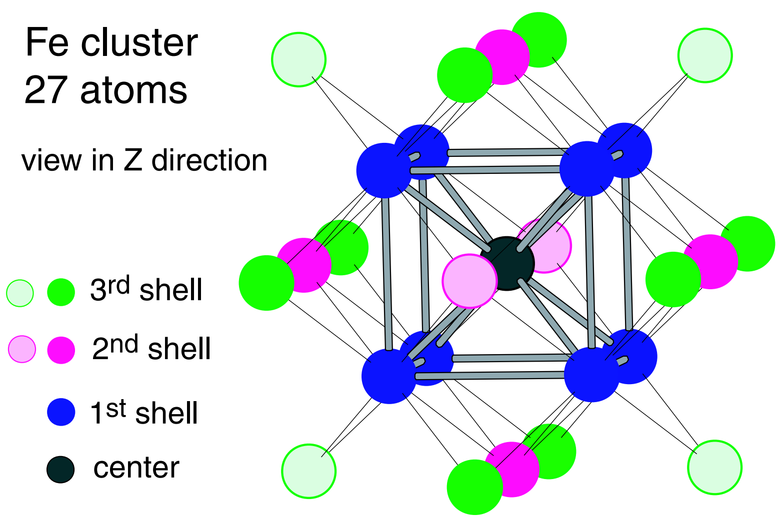
$\Rightarrow$ intuition has a limited role

$\Rightarrow$ one has to perform calculations for the real stuff

Systems to be studied

- Free spherical Fe clusters, geometry taken as if cut from a *bcc* Fe crystal
- Cluster sizes: between 9 atoms (1 coordination shell) and 89 atoms (7 coordinations shells)



shells	atoms	radius [a.u.]
1	9	4.70
2	15	5.42
3	27	7.67
4	51	8.99
5	59	9.39
6	65	10.85
7	89	11.82

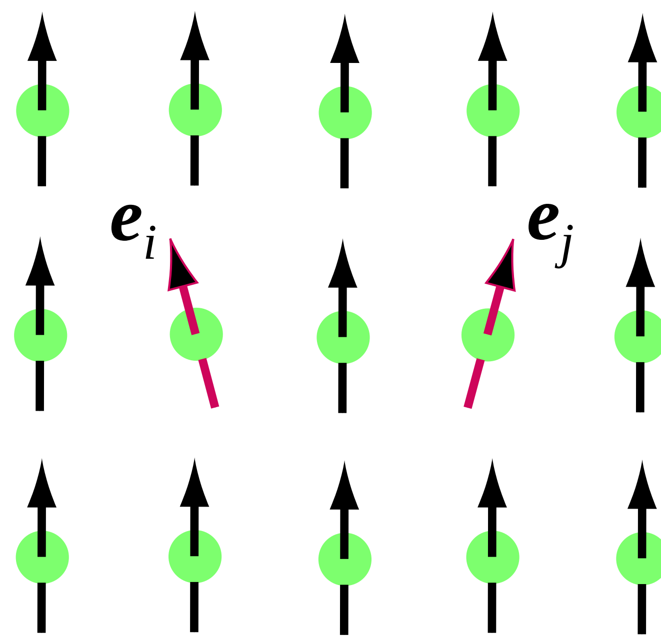
Calculations for $T = 0$

- *Ab-initio* within LDA framework (material specific)
- Scalar-relativistic real-space spin-polarized multiple-scattering formalism
- Atomic sphere approximation (ASA)
- Using SPRKKR code
  http://olymp.cup.uni-muenchen.de/ak/ebert/SPRKKR

Calculations for $T \neq 0$

For localized moments, finite-temperature magnetism can be described by a classical Heisenberg hamiltonian

$$H_{\mathrm{eff}} = -\sum_{i \neq j} J_{ij}\, \mathsf{e}_i \cdot \mathsf{e}_j$$



Mapping DFT onto Heisenberg

Coupling constants J_{ij} can be obtained from ground-state electronic properties:

$$J_{ij} \;=\; -\frac{1}{4\pi}\,\mathrm{Im}\int^{E_F}\mathrm{d}E\;\mathrm{Tr}\left[\,(t^{-1}_{i\uparrow}-t^{-1}_{i\downarrow})\;\tau^{ij}_{\uparrow}\;(t^{-1}_{j\uparrow}-t^{-1}_{j\downarrow})\;\tau^{ji}_{\downarrow}\,\right]$$

[Liechtenstein *et al.* JMMM **67**, 65 (1987)]

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Valid only if magnetism can be described by localized magnetic moments *(fine for Fe)*

From J_{ij} to $M(T)$

- Mean magnetization $M(T)$ of a system described by a classical Heisenberg hamiltonian is

$$M(T) = \frac{\sum_k M_k \exp(-\frac{E_k}{k_B T})}{\sum_k \exp(-\frac{E_k}{k_B T})}$$

M_k is the magnetization of the system for a particular configuration k of the directions of spins

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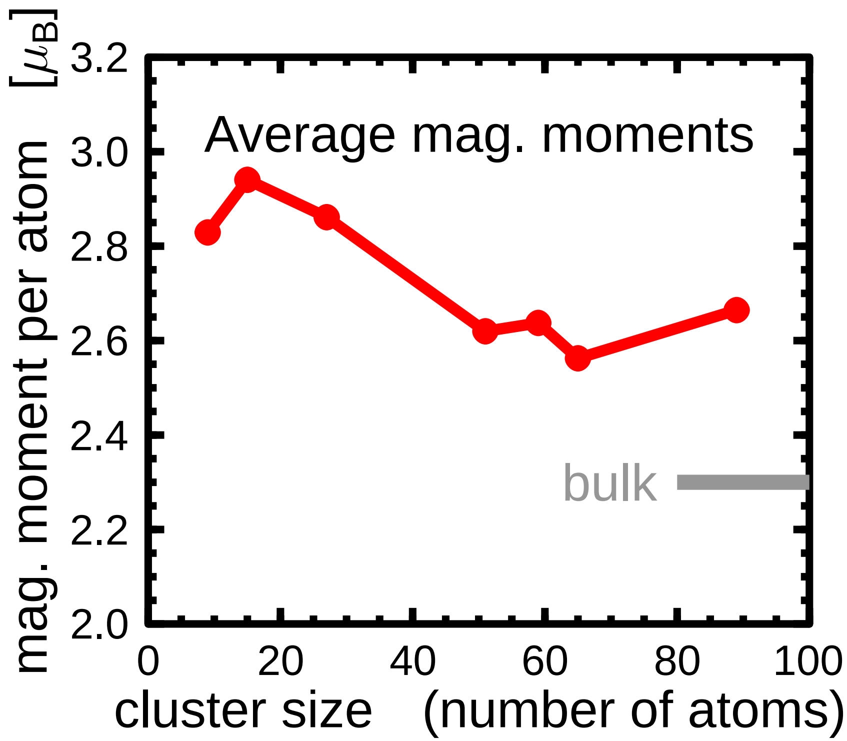
- For bulk Fe, this procedure yields finite-temperature results that are in a good agreement with experiment [Pajda *et al.* PRB **64**, 174402 (2001)]

Fe clusters at $T = 0$



- Average magnetic moment of clusters oscillates with cluster size

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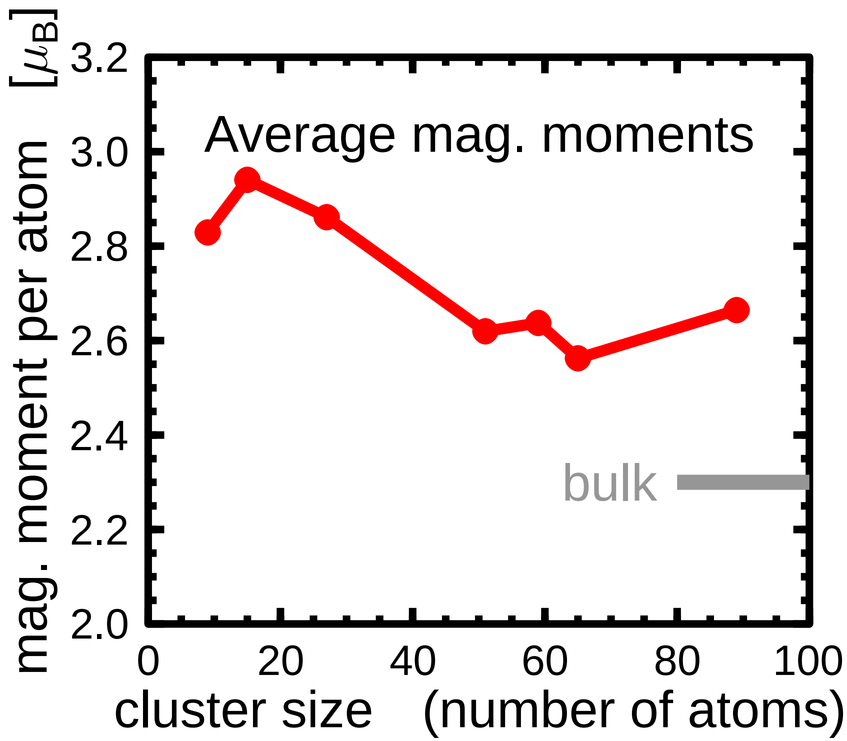

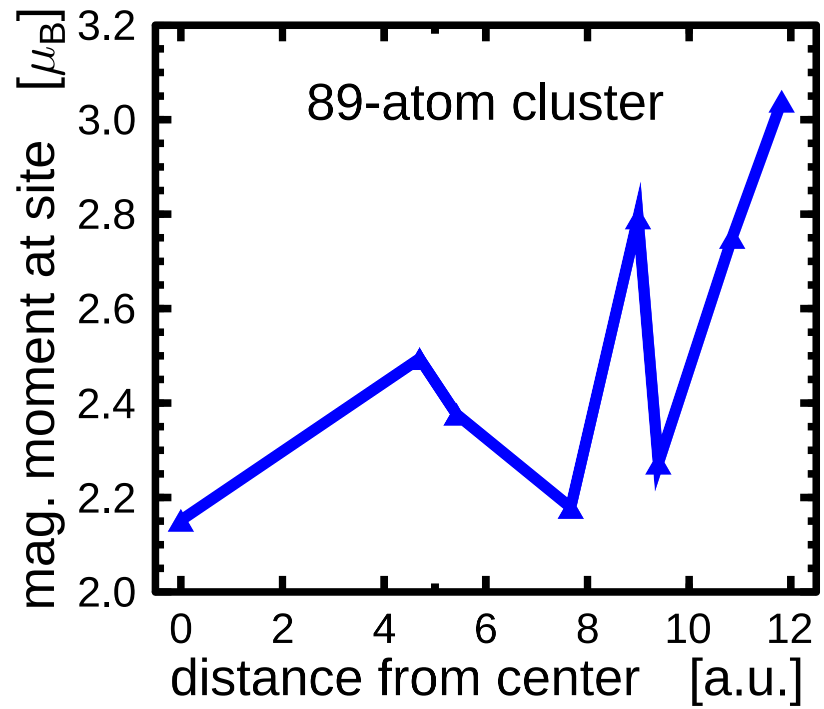

- Average magnetic moment of clusters oscillates with cluster size
- Local magnetic moments increase when going from the center outwards in an oscillatory way
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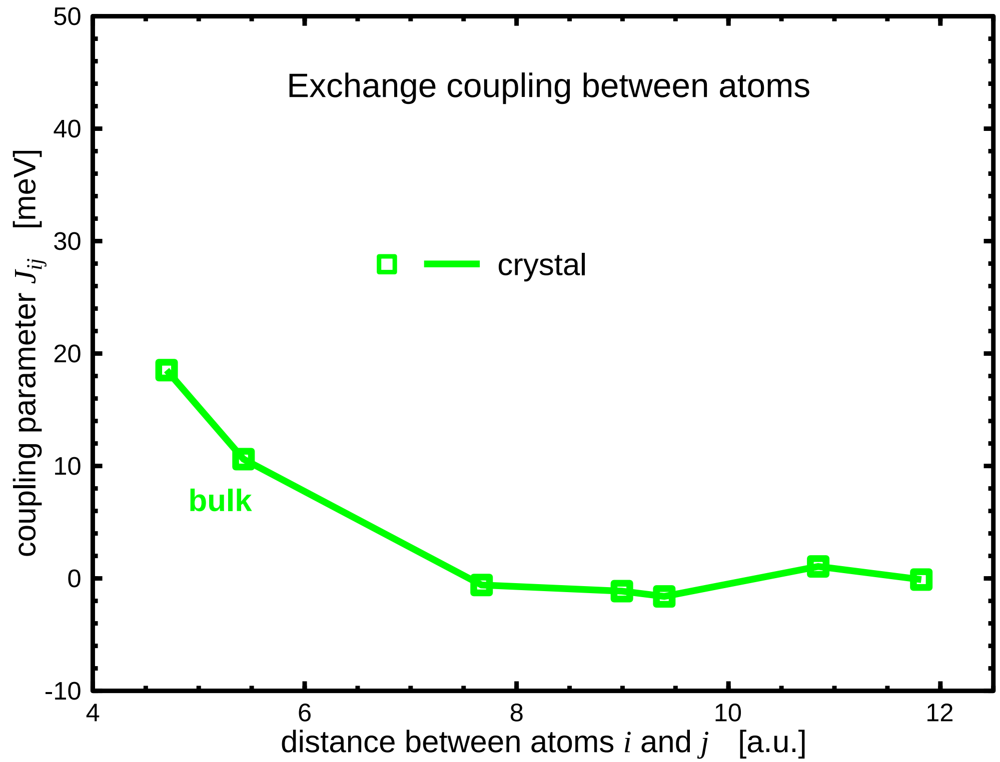
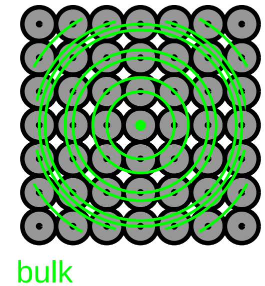
J_{ij} in bulk and in clusters





Atom i is fixed, atom j scans coordination shells around i

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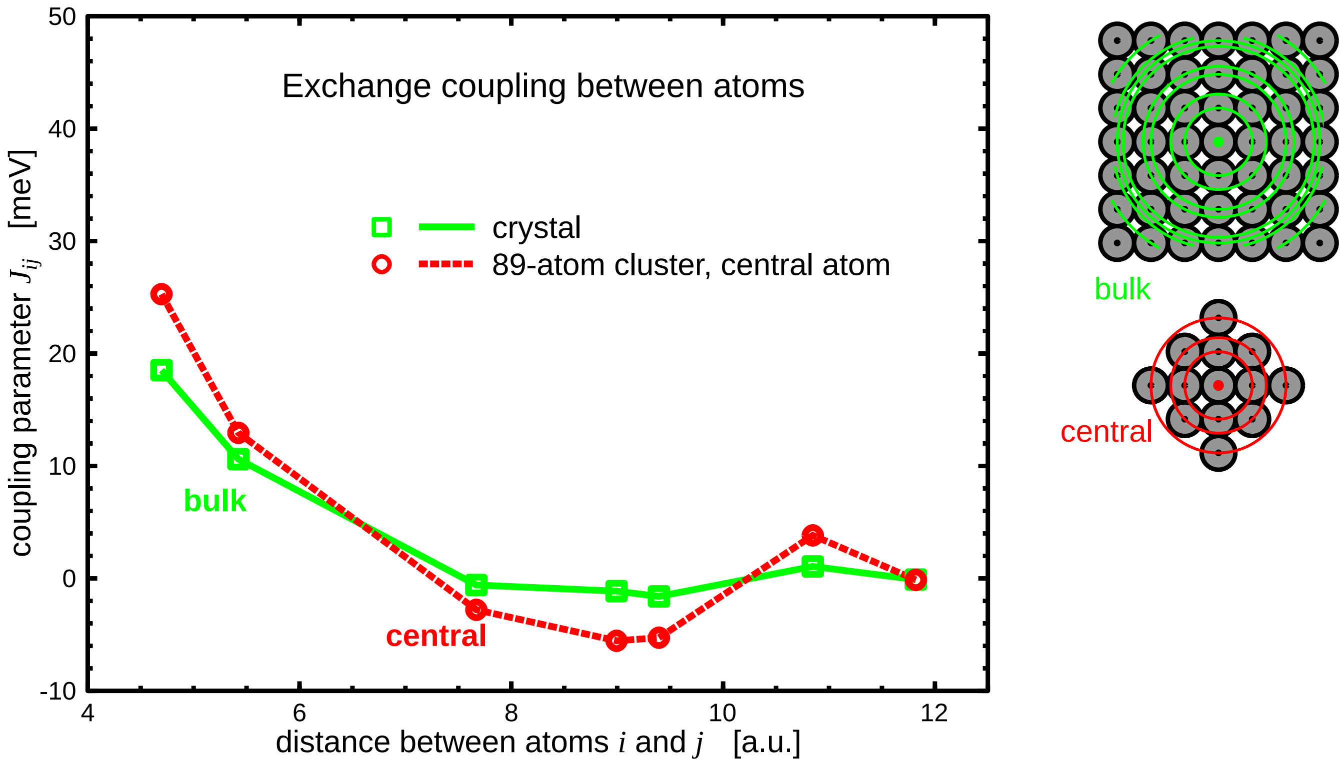

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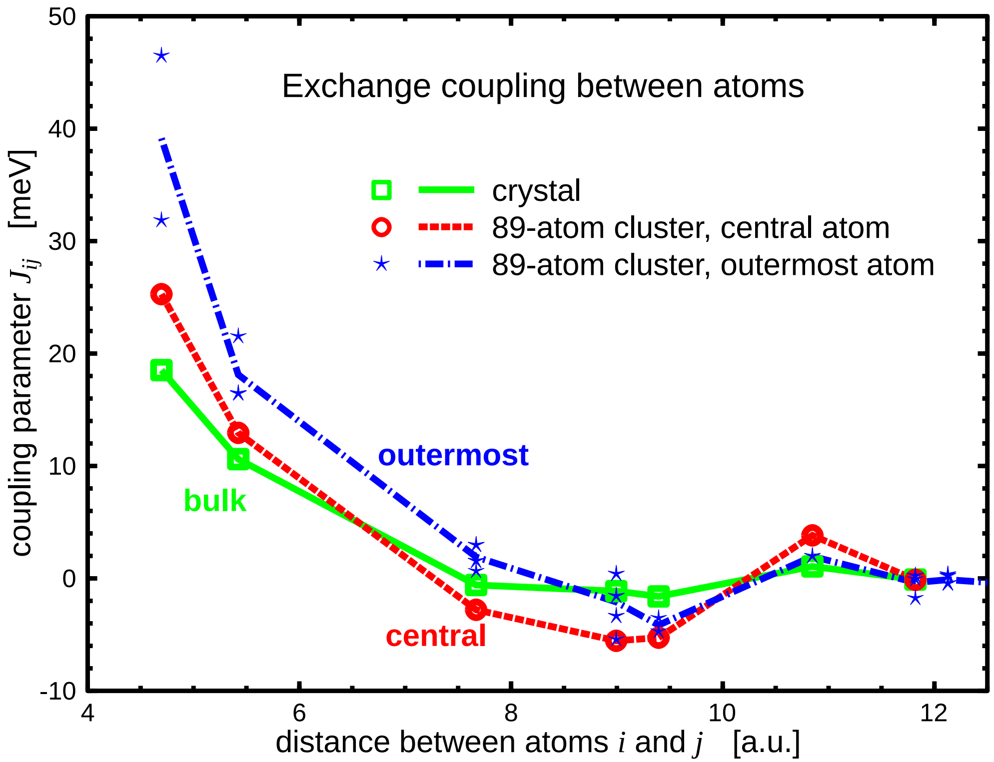

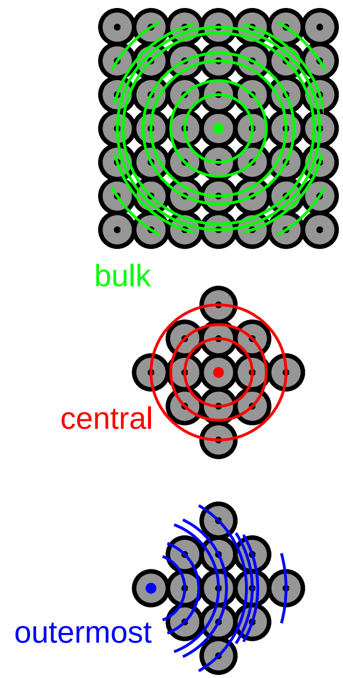

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- Oscillatory decay of J_{ij} with distance
- J_{ij} for a given distance differs a lot between clusters and crystals

Site-dependence of $\sum_j J_{ij}$

Total strength with which one spin (at site i) is held in its direction:

Energy needed to flip the spin of atom i while keeping all the remaining spins collinear:

$$J_i = \sum_{j \neq i} J_{ij}$$

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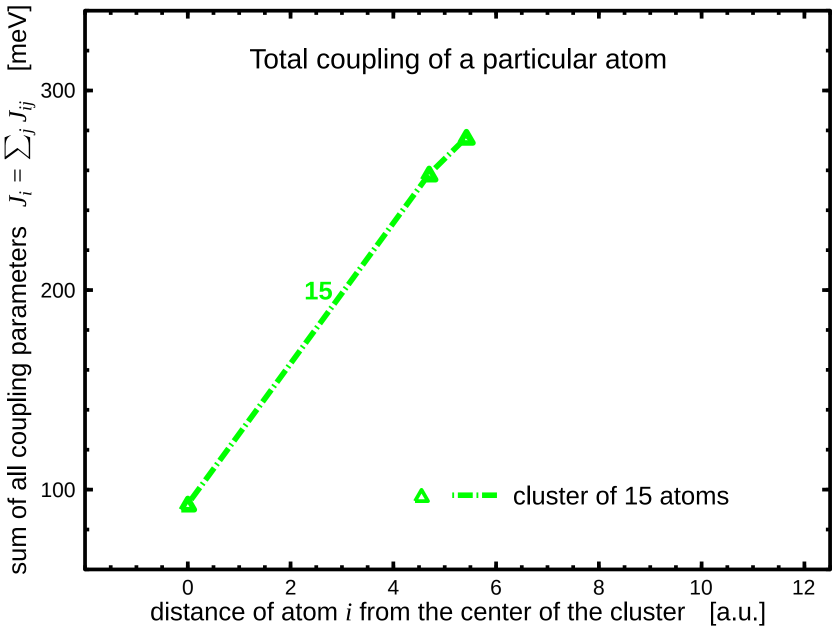

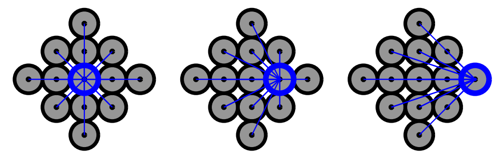

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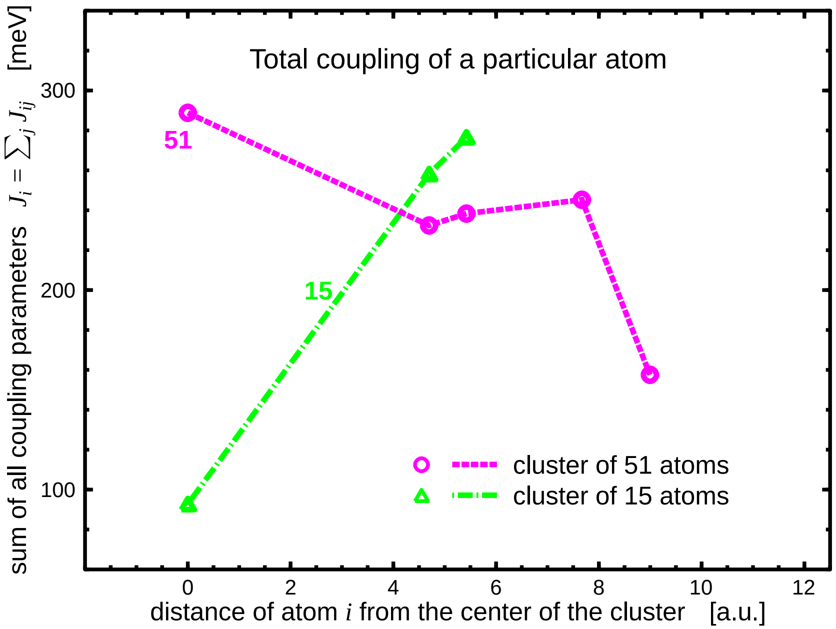

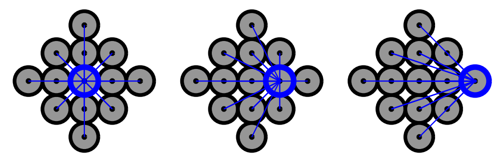

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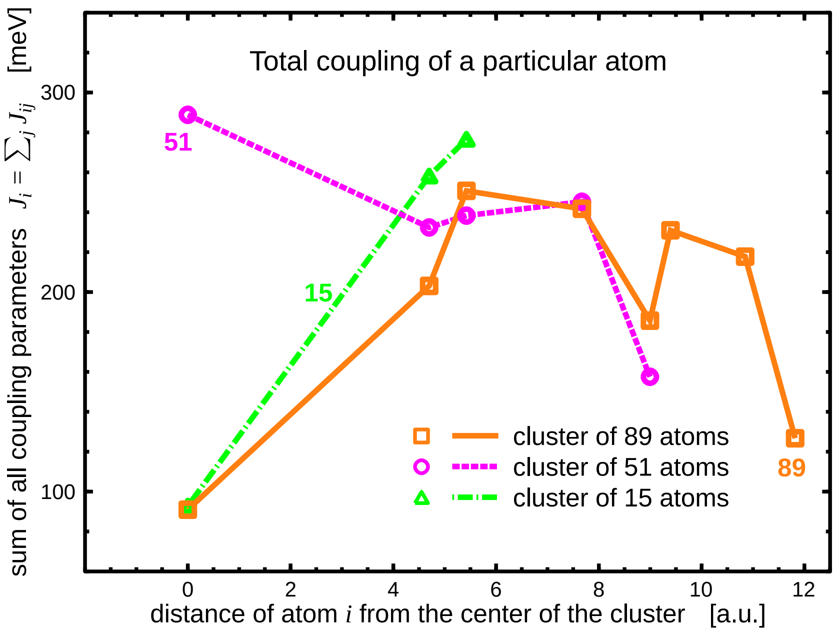

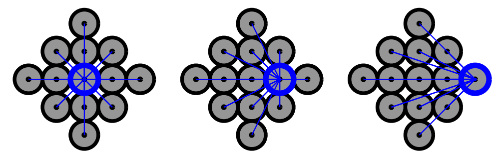

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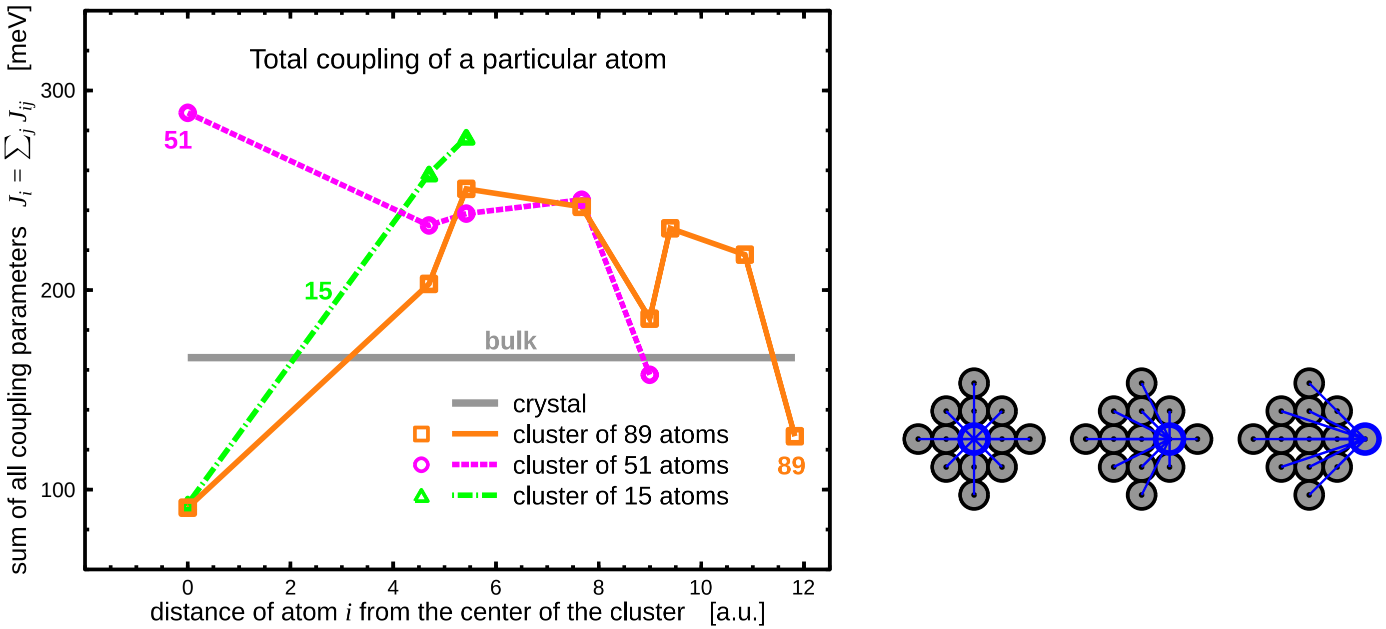
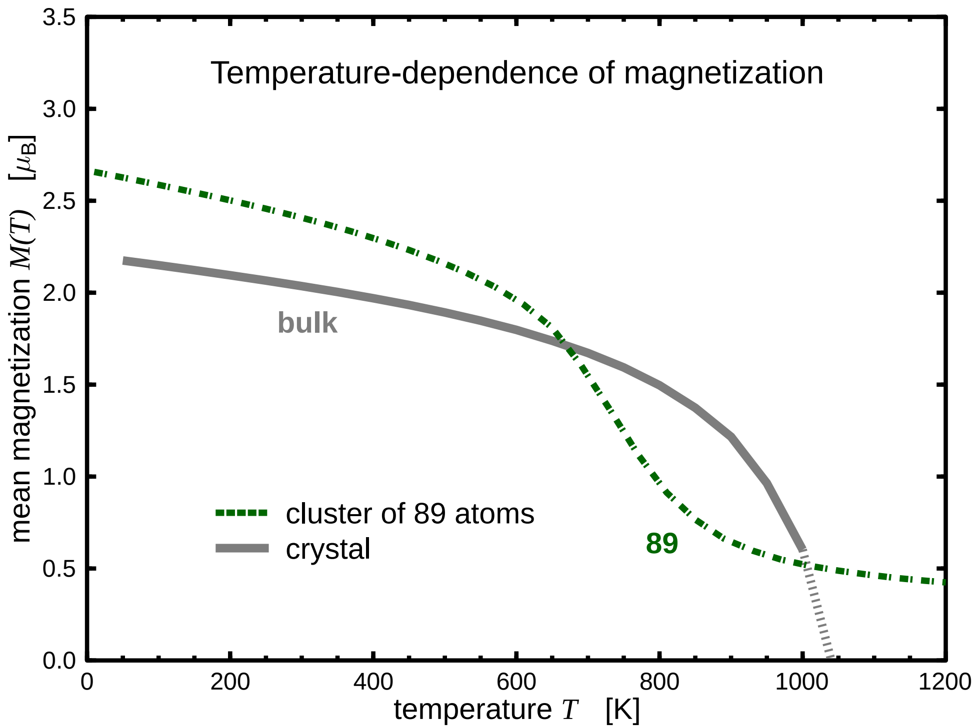


- Mild differences in J_{ij} translate into large differences in $\sum_j J_{ij}$
- No systematics in cluster size,
  no systematics in the position of atom within a cluster

Decay of magnetization with T



[Bulk $M(T)$ curve was extrapolated to calculated T_C]

- $M(T)$ curves are more shallow in clusters than in bulk

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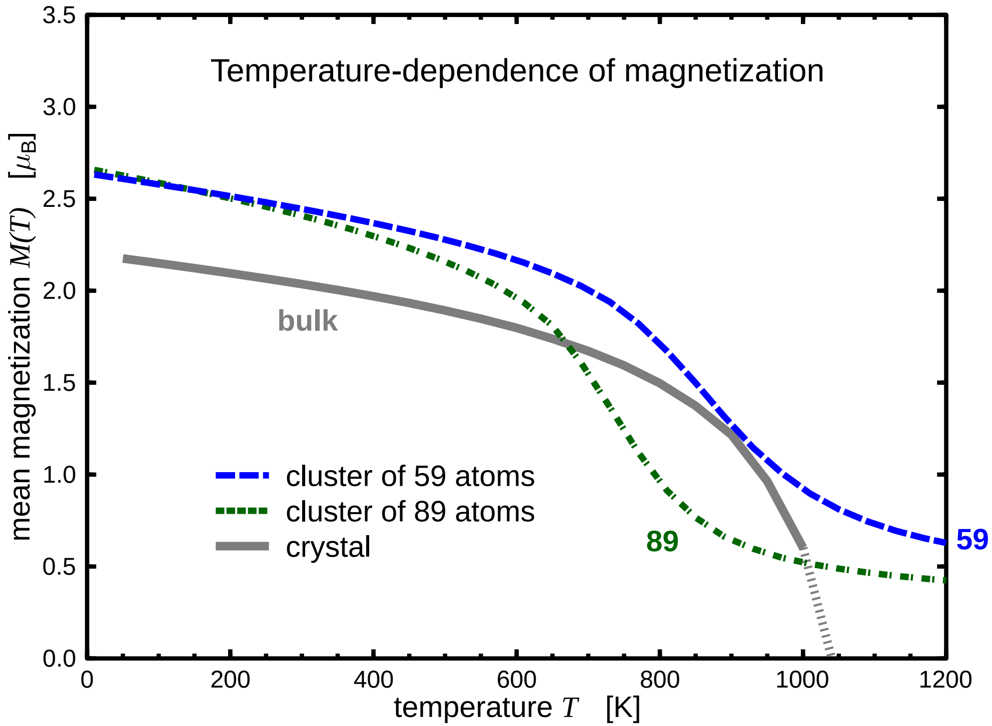

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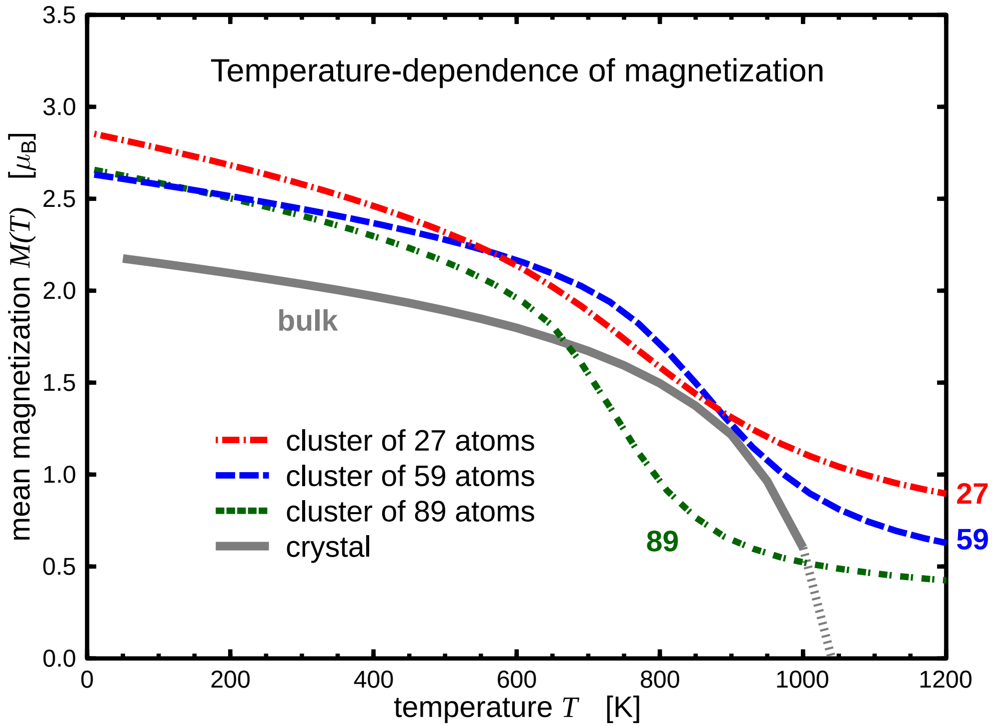

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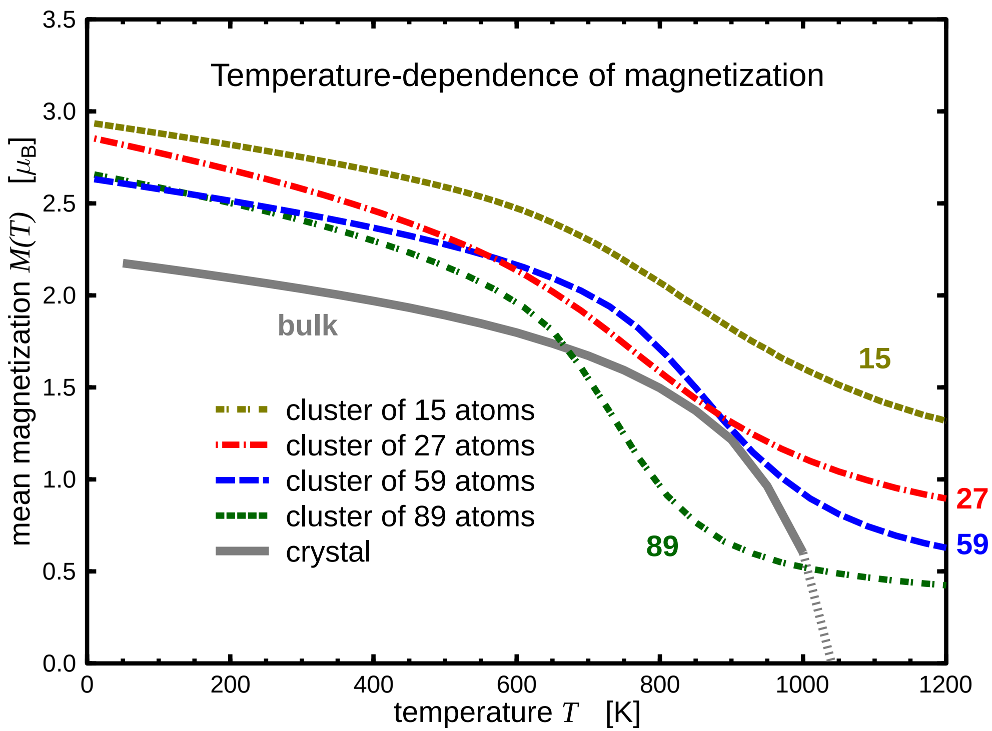

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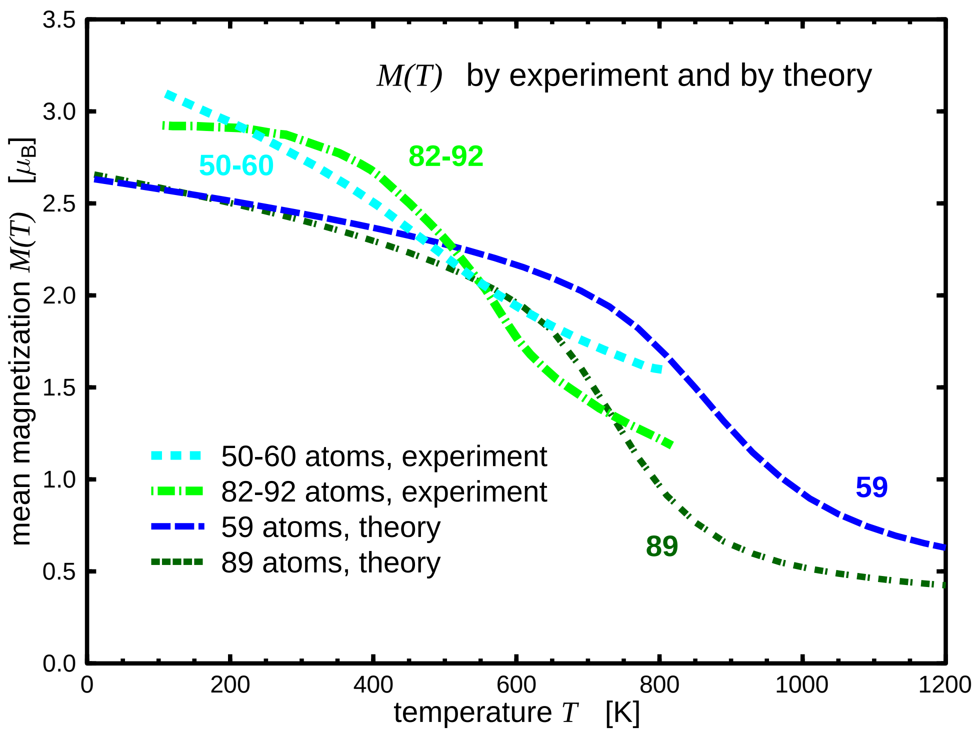
Experimental and theoretical $M(T)$

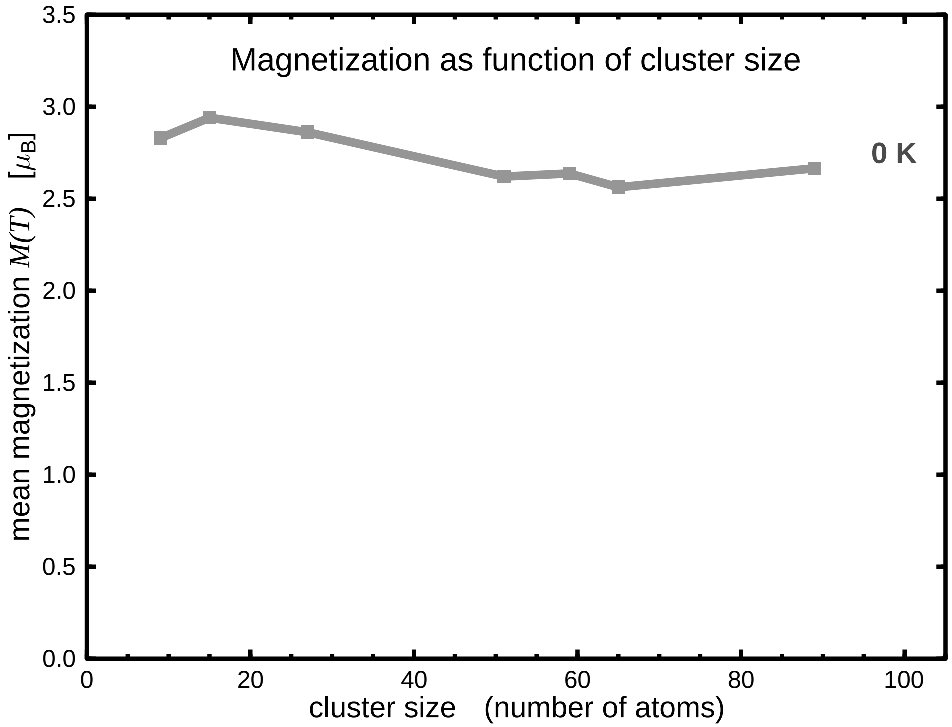


Experiment from Billas *et al.* PRL **71**, 4067 (1993)

Caution:

Each experimental curve corresponds to a *range of cluster sizes* (it represents an average over several $M(T)$ curves which may differ quite a lot one from another)

M as function of cluster size



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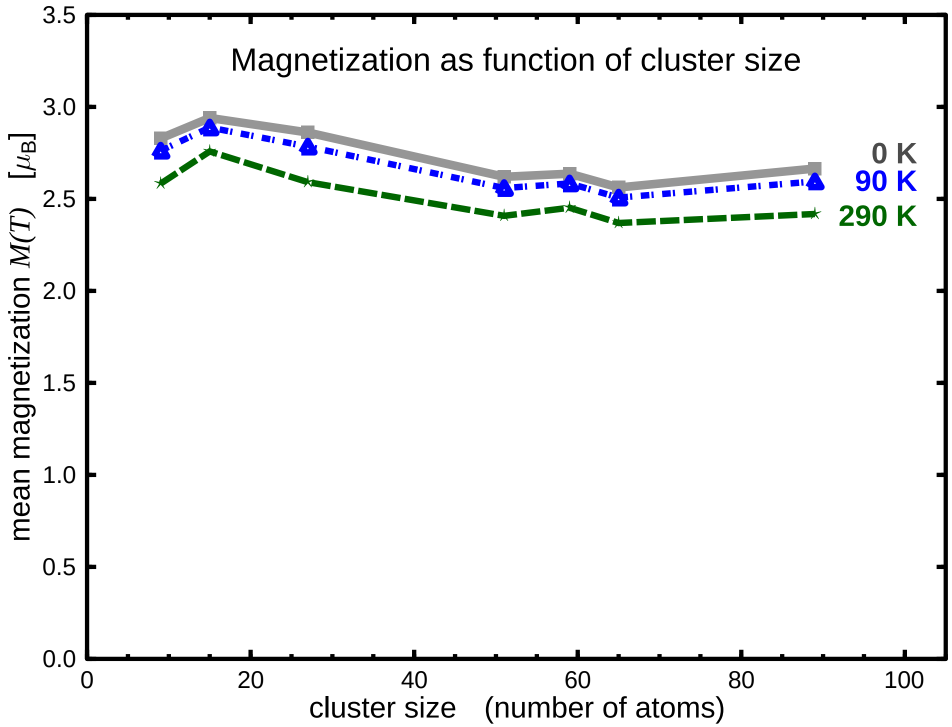

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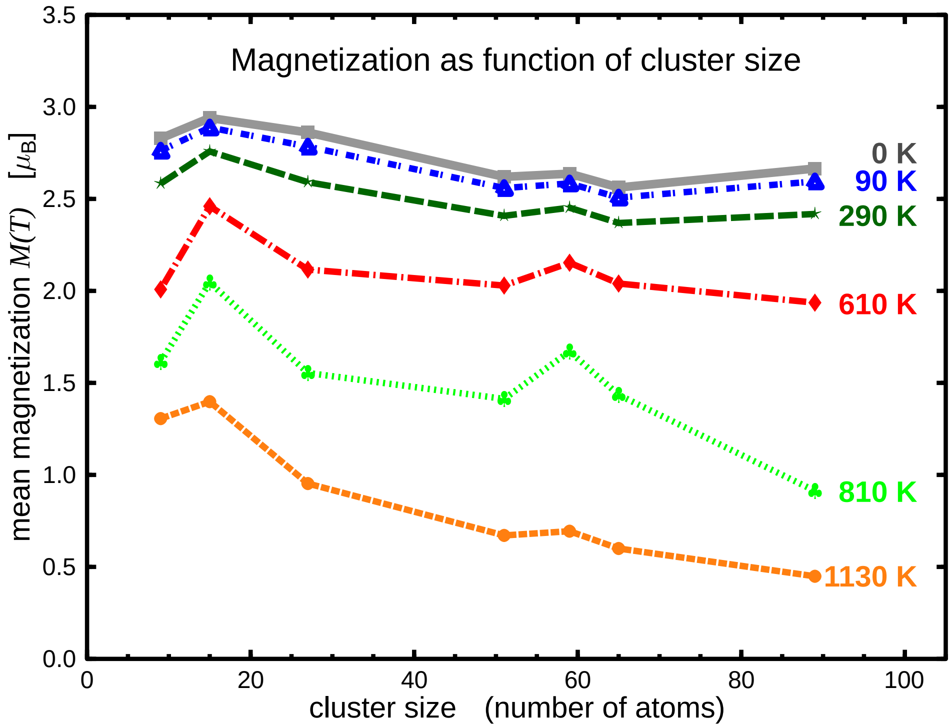

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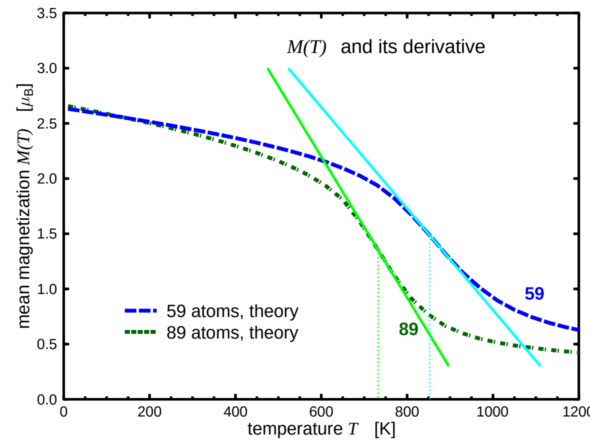
- For large T, magnetization of large clusters is significantly reduced

Dependence of T_c on cluster size



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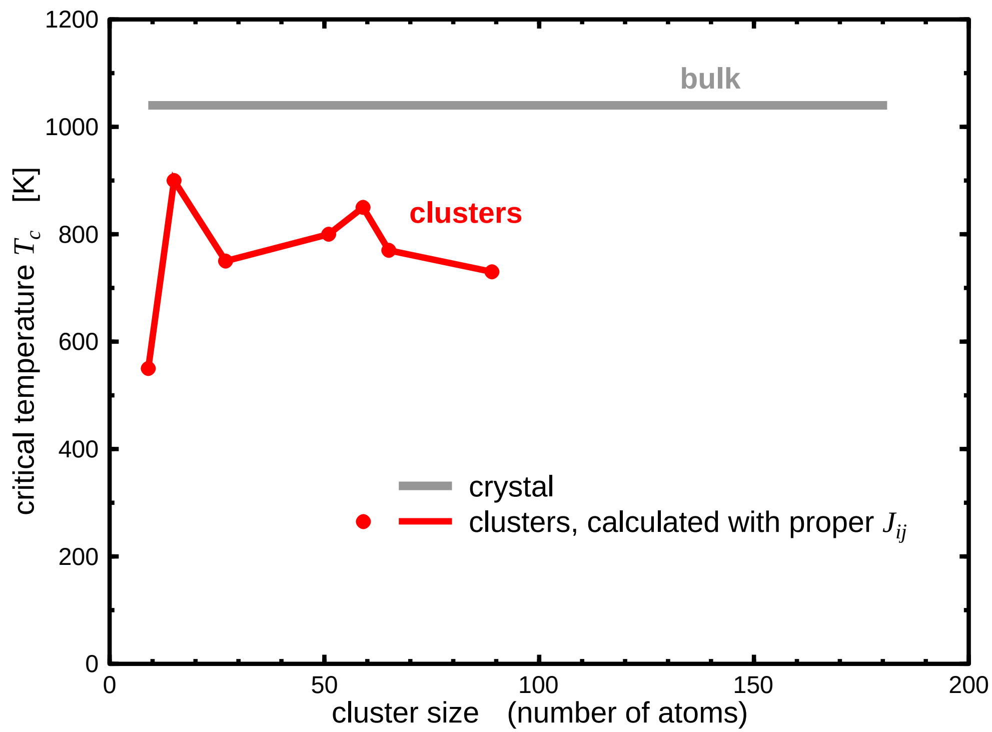

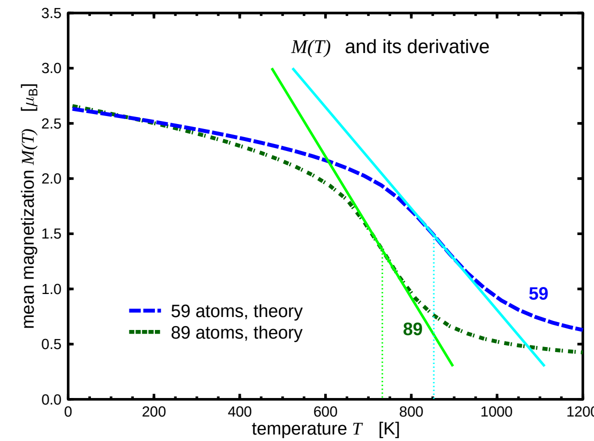

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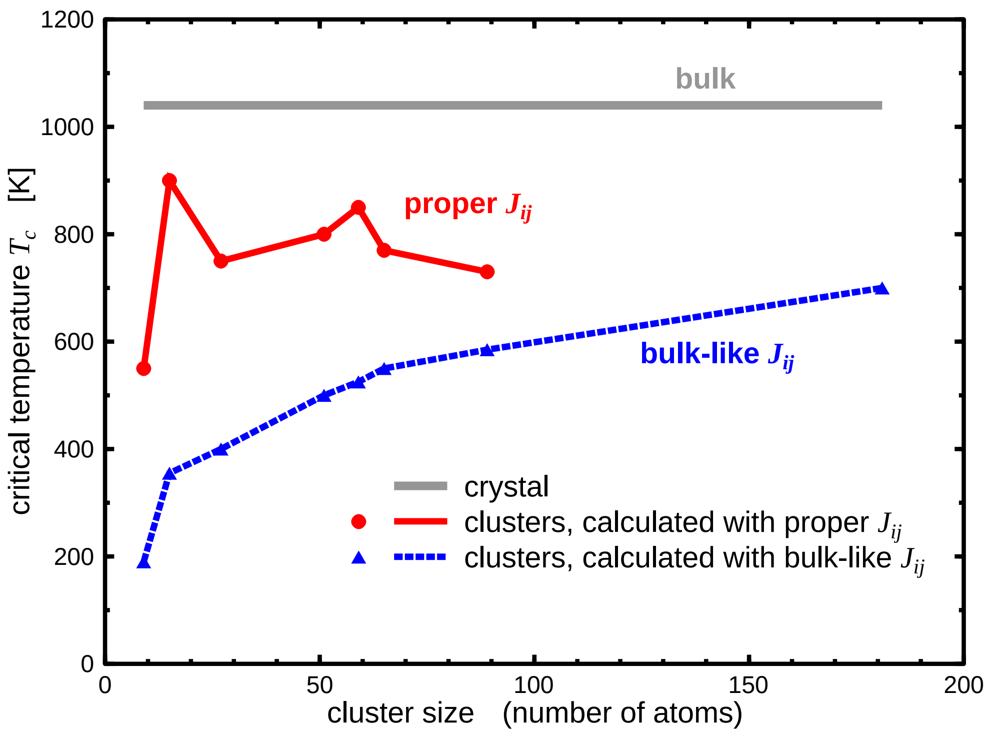

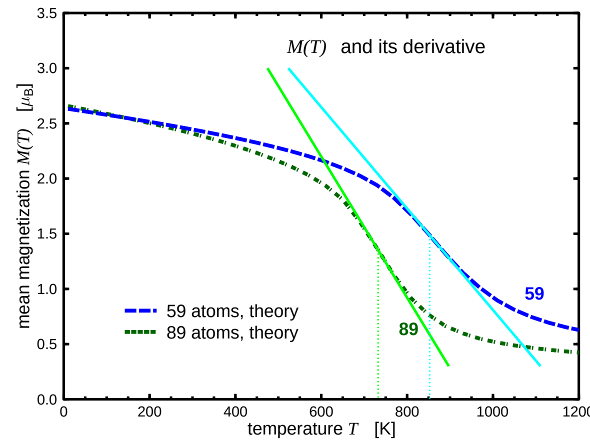

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- T_c oscillates with cluster size
- Proper cluster-adjusted J_{ij} have to be taken into account

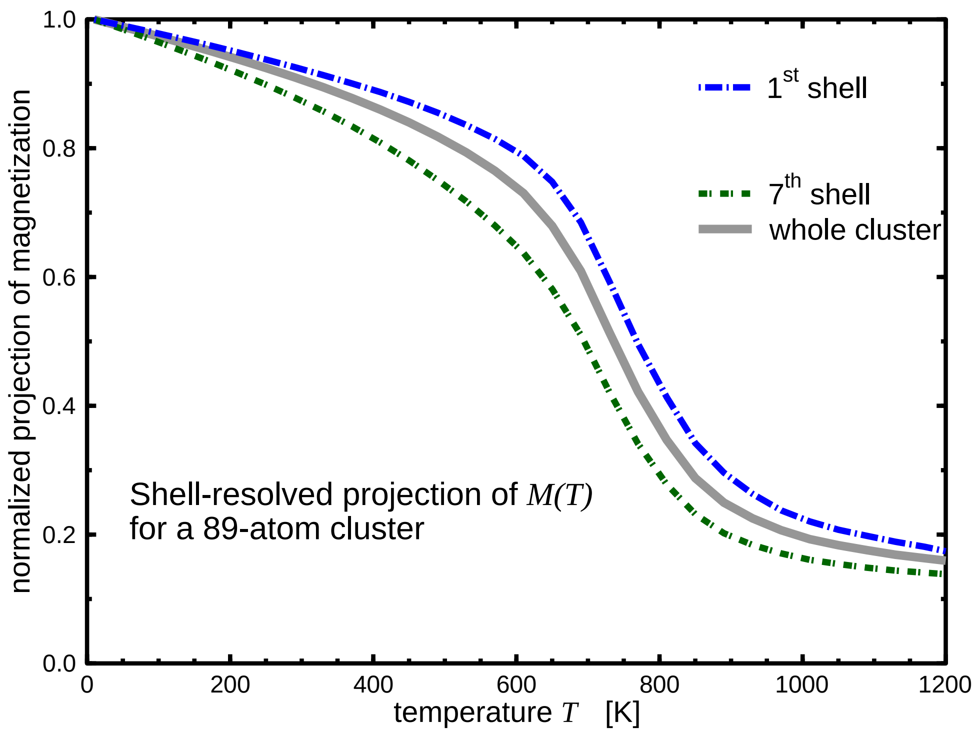
Shell-resolved magnetization

Expectations:

outer shells have smaller coordination numbers than inner shells

$\Rightarrow M$ in outer shells should decay more quickly with T than M of inner shells

Shell-resolved magnetization



Projecting M of a shell onto the direction of the total M of the 89-atom cluster

(Projections are normalized to $T = 0$)

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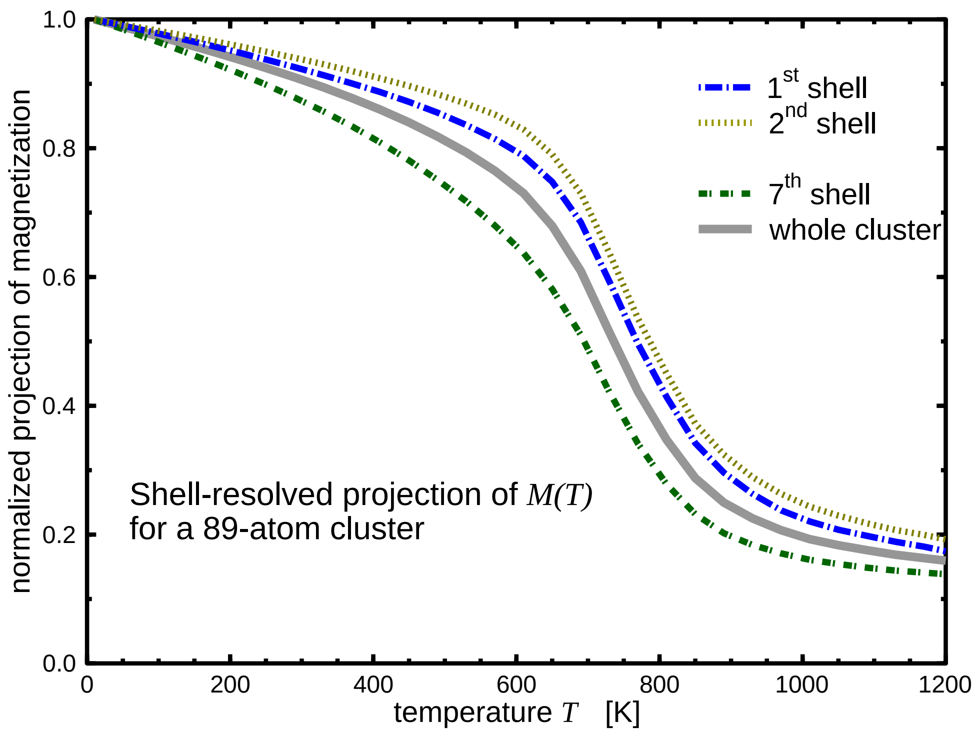

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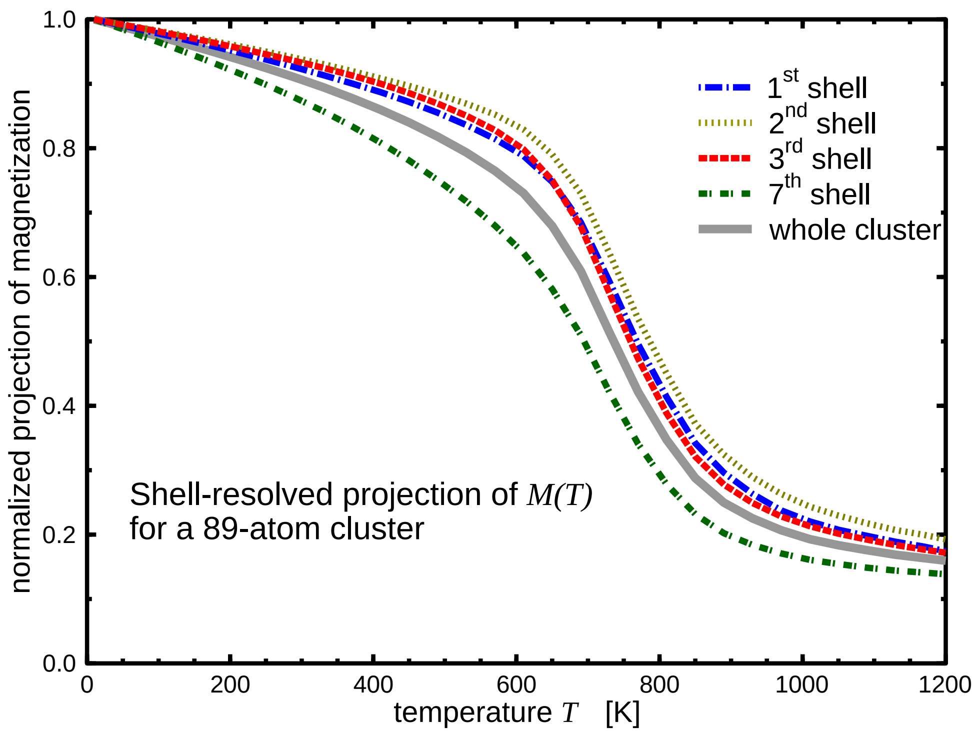

Projecting M of a shell onto the direction of the total M of the 89-atom cluster

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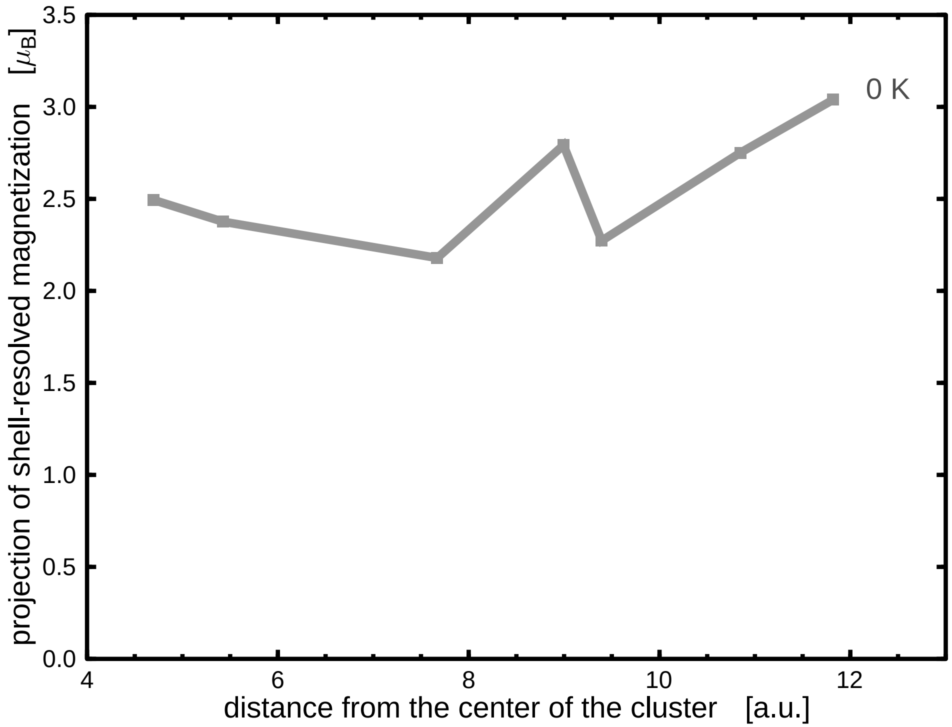
- Not monotonous in order of shells
- Although M of outer shells *usually* decays faster than M of inner shells, no systematics can be found.

Magnetic profile for $T \neq 0$



Cluster of 89 atoms
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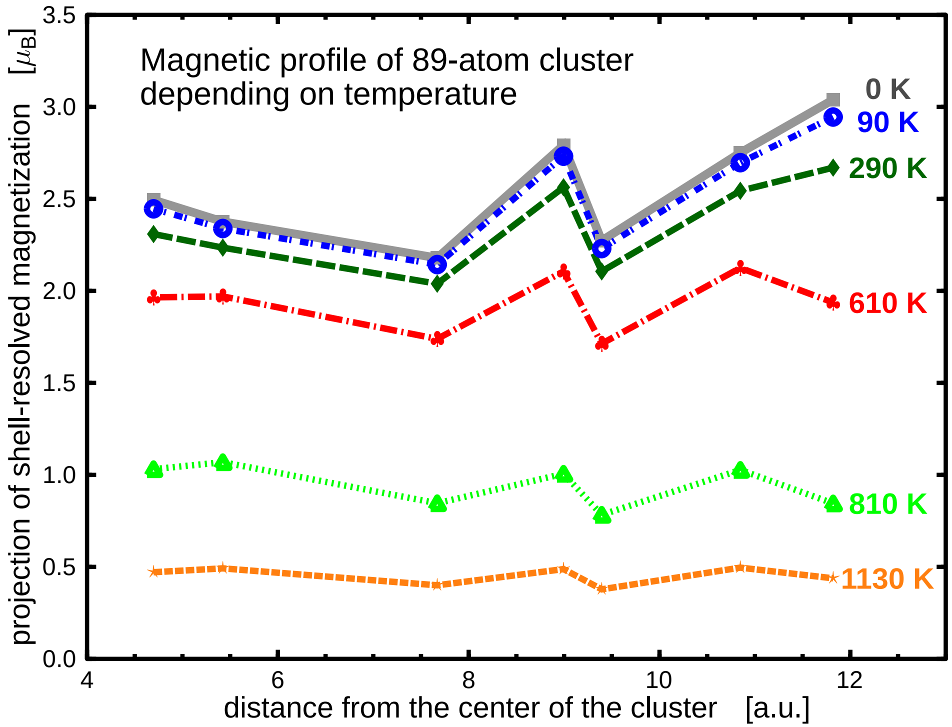

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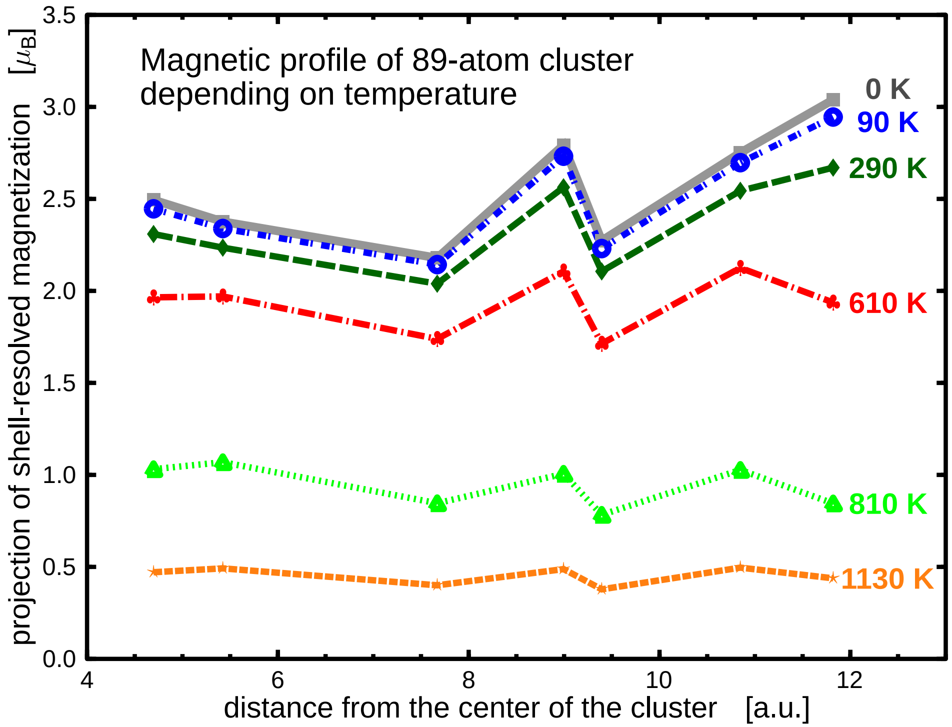

Cluster of 89 atoms
(7 coordination shells)

- Magnetic profile gets more flat if temperature increases
- M of outermost layers is similar in magnitude to M of inner layers even for large T (i.e., no drastic decrease of surface M)

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- For large T, magnetization of large clusters is significantly reduced
- Critical temperature T_c oscillates with cluster size
- (Normalized) M of the outer shells *usually* decreases with T more quickly than M of inner shells
- Simple models (such as taking J_{ij} from bulk) do not work, systematical trends cannot be guessed beforehand

  $\Rightarrow$ one really *has* to calculate all the quantities of interest