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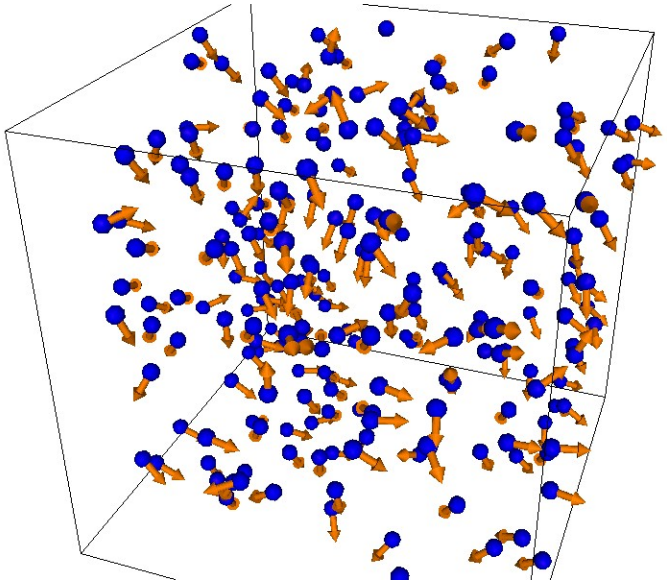
Atomistic Spin Dynamics

Simulations on Mn-doped GaAs and CuMn
Applications to spin wave instabilities



Ångströmlaboratoriet

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21 Apr 2009

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Outline

- Our approach to atomistic spin dynamics
- Benchmarks with Monte Carlo
- Details of equations of motion derivations
- Dynamics of dilute magnetic semiconductor Mn-doped GaAs
- Dynamics of the spin glass alloy CuMn
- Spin wave instabilities
- Outlook and Conclusions

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Different length scales in magnetization dynamics

Time dependent Spin-polarized Density Functional Theory (TD-SDFT)	Subatomic length scales, computationally heavy. Magnetization density as field $\mathbf{m}(\mathbf{r})$
Micromagnetics	Micron lengths, can simulate complete devices. magnetization density as field $\mathbf{m}(\mathbf{r})$

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Different length scales in magnetization dynamics

Time dependent Spin-polarized Density Functional Theory (TD-SDFT)	Subatomic length scales, computationally heavy. Magnetization density as field $\mathbf{m}(\mathbf{r})$
Atomistic Spin Dynamics	Nanometer length scales, magnetization density as discrete set of atomic magnetic moments $\mathbf{m}_i$
Micromagnetics	Micron lengths, can simulate complete devices. magnetization density as field $\mathbf{m}(\mathbf{r})$

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Motivations for use of atomistic spin dynamics for studies of dilute magnetic systems

- Investigate the magnetization temperature dependence in equilibrium; at varying dopant and defect concentrations.
- Investigate correlation functions, response functions etc.

This can be achieved with Monte Carlo simulations – what can be gained with atomistic spin dynamics?

- The (real) time evolution of the magnetization
- Time evolution of correlation functions
- Dynamic response functions

Spin dynamics studies on

Mn-doped GaAs: Hellsvik et al., PRB **78**, 144419 (2008)
The spin glass alloy CuMn : Skubic et al., PRB **79**, 024411 (2009)

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Equations of motion: Atomistic Landau-Lifshitz Equation

$$\frac{d\mathbf{m}_i}{dt} = -\gamma \mathbf{m}_i \times [\mathbf{B}_i + \mathbf{b}_i(t)] - \gamma \frac{\alpha}{m} \mathbf{m}_i \times (\mathbf{m}_i \times [\mathbf{B}_i + \mathbf{b}_i(t)])$$

Precession term

Damping term

$\mathbf{m}_i$ atomic magnetic moment

γ gyromagnetic ratio

$\mathbf{B}_i$ effective magnetic field

α damping term

$\mathbf{b}_i(t)$ stochastic magnetic field

Magnetic exchange is mapped onto a Heisenberg Hamiltonian $\mathscr{H}_{\mathrm{ex}} = -\frac{1}{2} \sum_{i \neq j} J_{ij} \mathbf{m}_i \cdot \mathbf{m}_j$

Derived from spin-polarized KS-Hamiltonian following V. P. Antropov et al. PRB **54**, 1019 (1996).

Our approach and implementation described in B. Skubic et al. JPhys: Cond Mat **20**, 315203 (2008).

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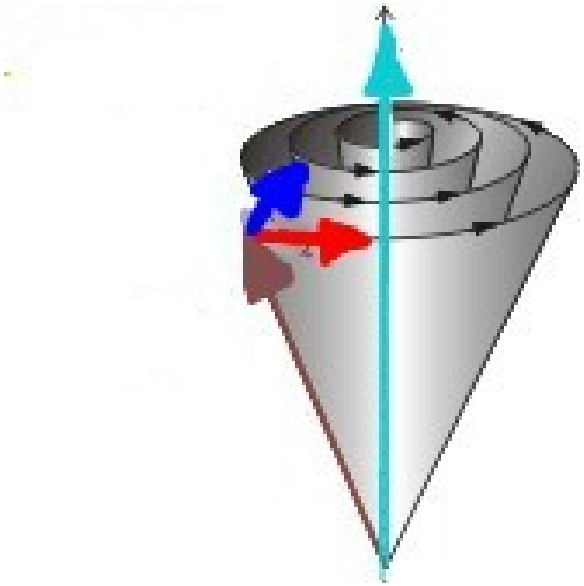
Equations of motion: Atomistic Landau-Lifshitz Equation

$$\frac{d\mathbf{m}_i}{dt} = -\gamma \mathbf{m}_i \times [\mathbf{B}_i + \mathbf{b}_i(t)] - \gamma \frac{\alpha}{m} \mathbf{m}_i \times (\mathbf{m}_i \times [\mathbf{B}_i + \mathbf{b}_i(t)])$$

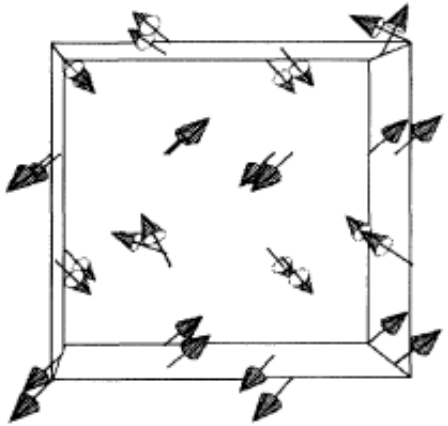
Precession term

Damping term



Precession of a single atomic moment in the effective magnetic field.

Simulation cells typically have periodic boundary conditions.



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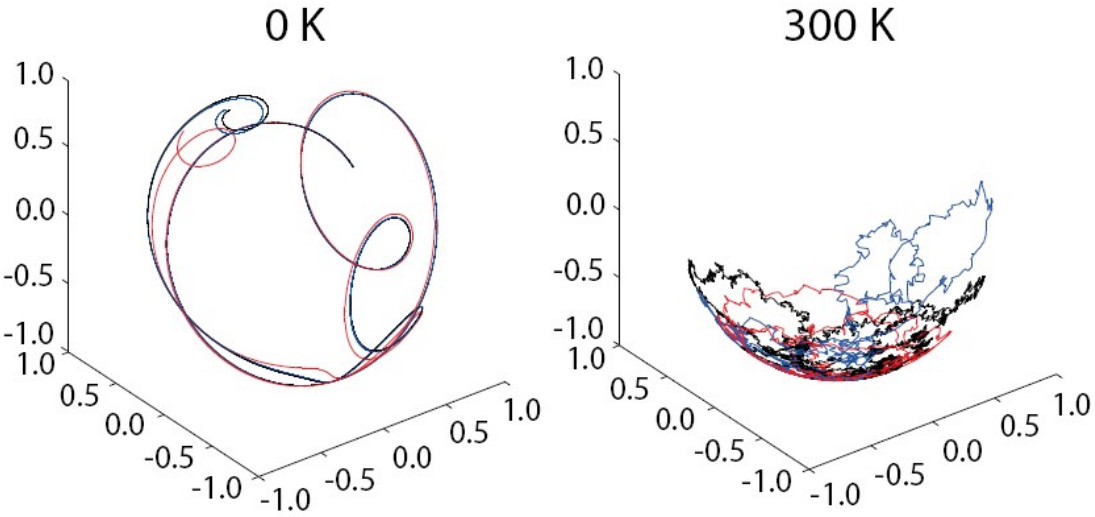
Trajectories at T=0 K resp 300 K



Δt:
10^{-18} s
10^{-17} s
10^{-16} s

Finite temperature effects are treated within a Langevin dynamics approach. The fluctuating magnetic field is normal distributed with an amplitude related to the damping parameter.

$$\langle b_{i,\mu}(t) \rangle = 0$$

$$\langle b_{i,\mu}(t) b_{j,\nu}(s) \rangle = 2D\delta_{\mu\nu}\delta_{ij}\delta(t-s)$$

$$D_{SLLG} = \frac{\alpha}{1+\alpha^2}\frac{k_B T}{\gamma m}$$

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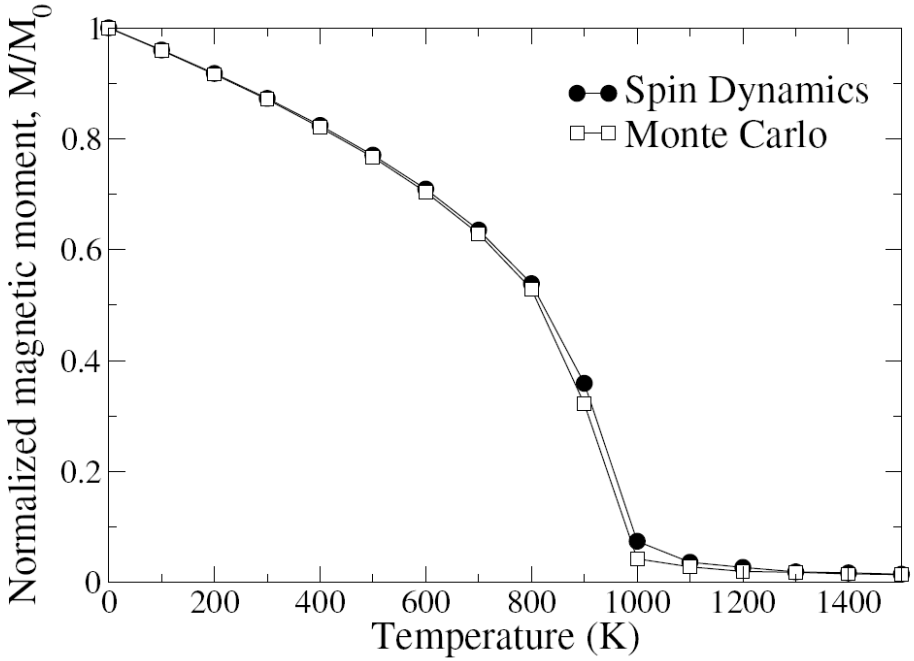
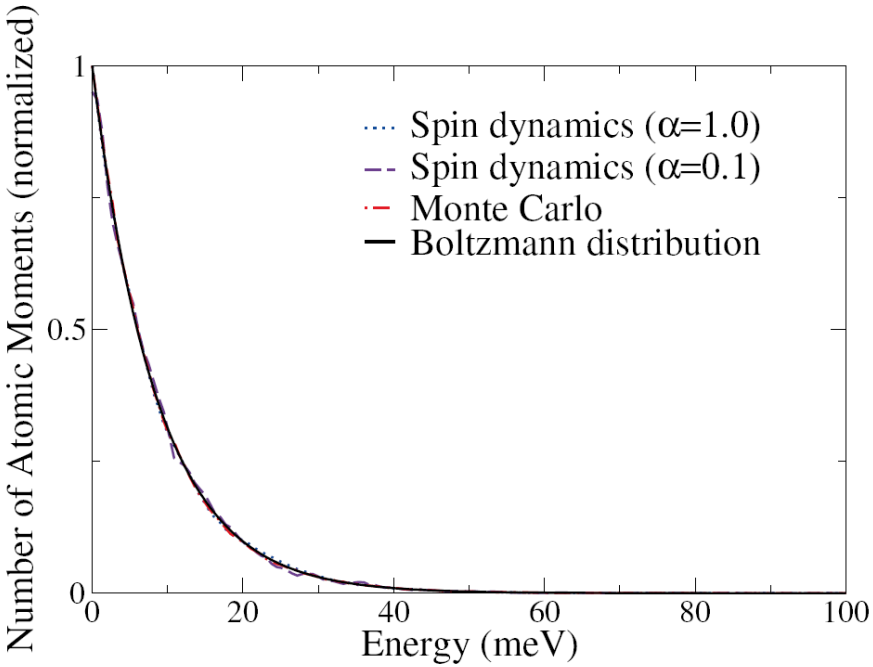
The ensemble average of the magnetization

The equilibrium configurations conform with those obtained in Monte Carlo simulations.





The distribution of magnetic moment energies.

Simulations are on bcc Fe, 4 exchange shells from DFT calc.

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Primary variables of TD-SDFT

$$n(\mathbf{r},t) = \sum_i \langle \Psi | \delta(\mathbf{r}-\mathbf{r}_i) | \Psi \rangle$$

Particle density

$$\mathbf{m}(\mathbf{r},t) = \mu_B \sum_i \langle \Psi | \boldsymbol{\sigma} \delta(\mathbf{r}-\mathbf{r}_i) | \Psi \rangle$$

Magnetization density

$$\mu_B = \frac{q\hbar}{2mc}$$

$$\Psi = \Psi(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N, t)$$

Derivation of equations of motion from the spin-polarized Kohn-Sham Hamiltonian

$$\hat{H}^{KS} = \sum_i \left[-\frac{\hbar^2 \hat{\nabla}_i^2}{2m} + v_s(\mathbf{r},t) - \mu_B \boldsymbol{\sigma} \cdot \mathbf{B}_s(\mathbf{r},t) \right]$$

$$\hat{H}^{KS}\Phi = i\hbar \frac{\partial \Phi}{\partial t}$$

$$\mathbf{B}_s(\mathbf{r},t) = \mathbf{B}(\mathbf{r},t) + \mathbf{B}_{xc}(\mathbf{r},t)$$

Current operator $$\hat{\mathbf{j}}(\mathbf{r},t) = \frac{\hbar}{2mi}\sum_i^N \nabla_i\delta(\mathbf{r}-\mathbf{r}_i) + \delta(\mathbf{r}-\mathbf{r}_i)\nabla_i$$

KS spin-current $$J^{KS}(\mathbf{r},t) = \mu_B \sum_i \langle\Phi|\boldsymbol{\sigma}\otimes\mathbf{j}_i(\mathbf{r},t)|\Phi\rangle$$

Evaluate the commutator of the magnetic operator and the KS-Hamiltonian

$$\frac{d\mathbf{m}(\mathbf{r},t)}{dt} = \frac{i}{\hbar}\langle\Phi|[H^{KS},\mathbf{m}(\mathbf{r},t)]|\Phi\rangle$$

The magnetic operator commutes with the scalar potential but not with the magnetic potential or the kinetic energy (non-relativistic case)

$$\frac{d\mathbf{m}(\mathbf{r},t)}{dt} + \hat{\nabla}\cdot J^{KS}(\mathbf{r},t) = \frac{q}{2mc}\mathbf{m}(\mathbf{r},t)\times\mathbf{B}_s(\mathbf{r},t)$$

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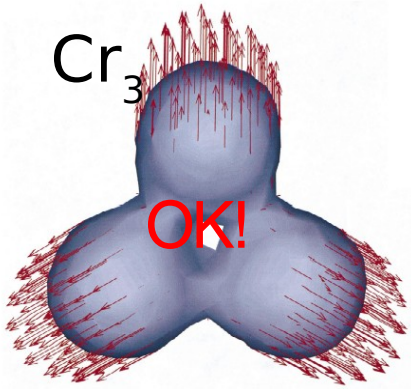
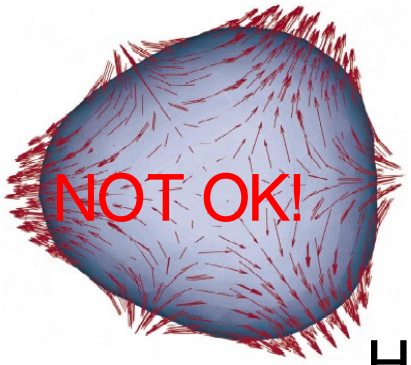
$$\frac{d\mathbf{m}(\mathbf{r},t)}{dt} + \hat{\nabla}\cdot J^{KS}(\mathbf{r},t) = \frac{q}{2mc}\mathbf{m}(\mathbf{r},t)\times\mathbf{B}_s(\mathbf{r},t)$$

Restrict to systems where:
1. rigid atomic moments
2.. spin currents can be discarded

$$\hat{\nabla}\cdot J^{KS}(\mathbf{r},t) = 0$$





But in standard LDA (or GGA): $\mathbf{m}(\mathbf{r},t) \parallel \mathbf{B}_{xc}(\mathbf{r},t)$
- and no torque would drive the magnetization!

Remedies? Depart from strict LDA

Map to Heisenberg Hamiltonian (our approach)

Construct new xc-Functionals (OEP, EXX)

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Other approaches to atomistic spin dynamics

Constrained local moments model (Stocks)
(builds on constrained DFT)

Spin cluster expansion (Fähnle)
(Extension of cluster expansion techniques in alloy theory)

Hybrid MC-SD (Landau)
Thermalization of spin system with Monte Carlo simulation, thereafter spin dynamics evolution at zero Temperature)

(this listing is by no means complete!)

Stocks et al., Phil. Mag **78**, 665 (1998)
Fähnle et al., Comp. Mat. Sci. **32**, 118 (2005)
Tao, Landau et al., Phys. Rev. Lett **95**, 087207 (2005)

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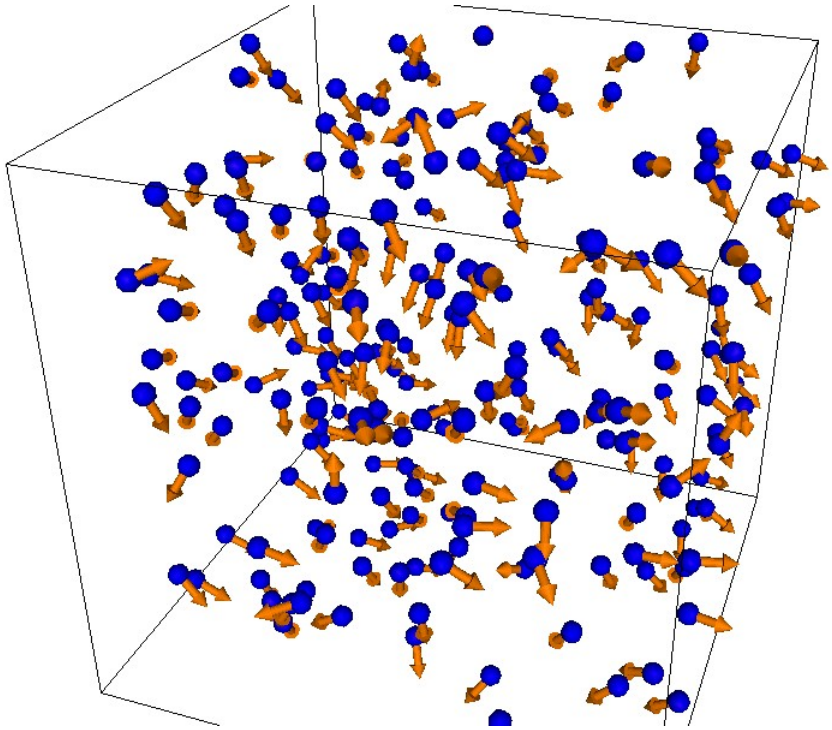
Application: SD Simulation of Mn-doped GaAs

Lattice structure: Zincblende with 5% of Ga atoms substituted with Mn atoms. Varying Arsenide antisite concentration. This screenshot for T=100 K and no antisites

Only Mn atoms are shown in visualization



Mn-doped GaAs: Hellsvik et al., PRB **78**, 144419 (2008)

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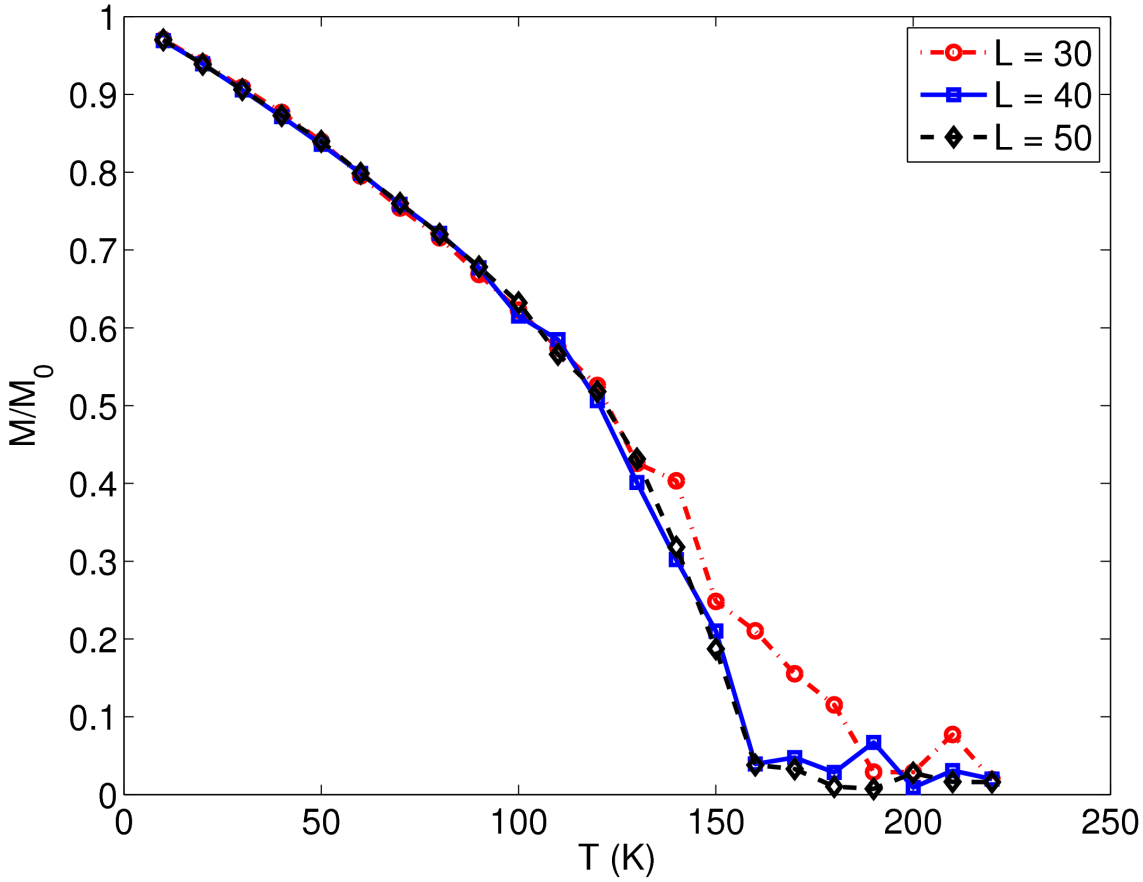
What size of simulations cell is necessary?



Different cell sizes: L=30, 40, 50

(i.e. 5400, 12800 or 25000 Mn atoms)

Same J_ij exchange values as in
J. Kudrnovský et al. PRB **69**, 115208 (2004)
L. Bergqvist et al. PRL **93**,137202, PRB **72**, 195210

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Time evolution of the magnetic order parameter

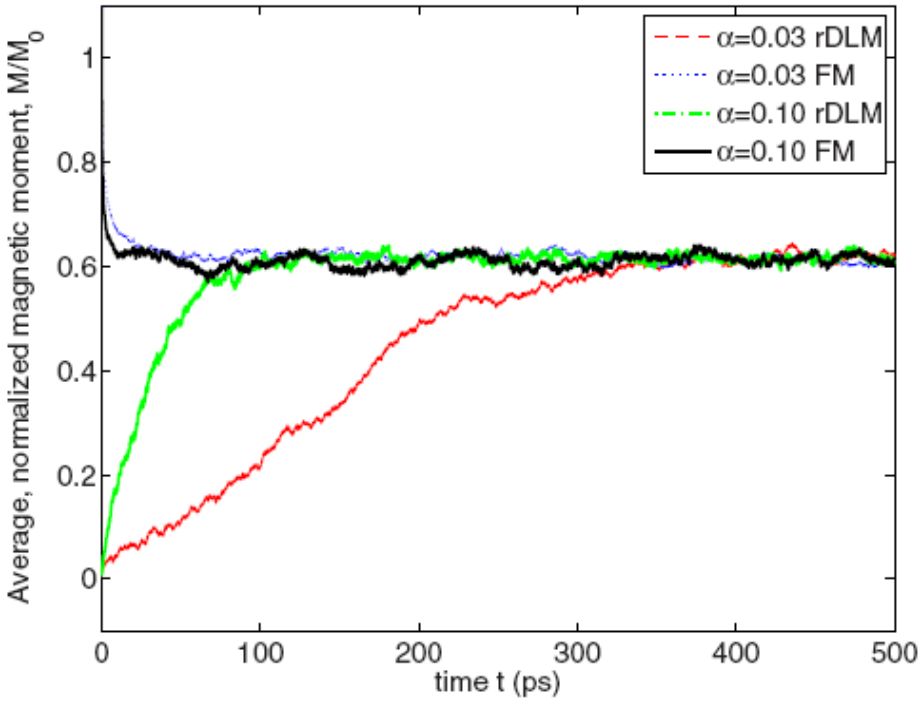

FIG. 4. (Color online) Time evolution of the average normalized magnetization starting from ferromagnetic (blue) and random (red) spin configurations for $L=40$, with As Antisite concentration $y=0.25\%$ at temperature $T=100$ K and with a damping parameter of 0.03. Similar simulations but with a damping parameter of 0.1 are shown in green and black for the random and ferromagnetic configurations, respectively.

The time scale is affected by:

Cell size (as for MC simulations)
T / Tc ratio, crit. slowing down (as for MC simulations)
The damping parameter

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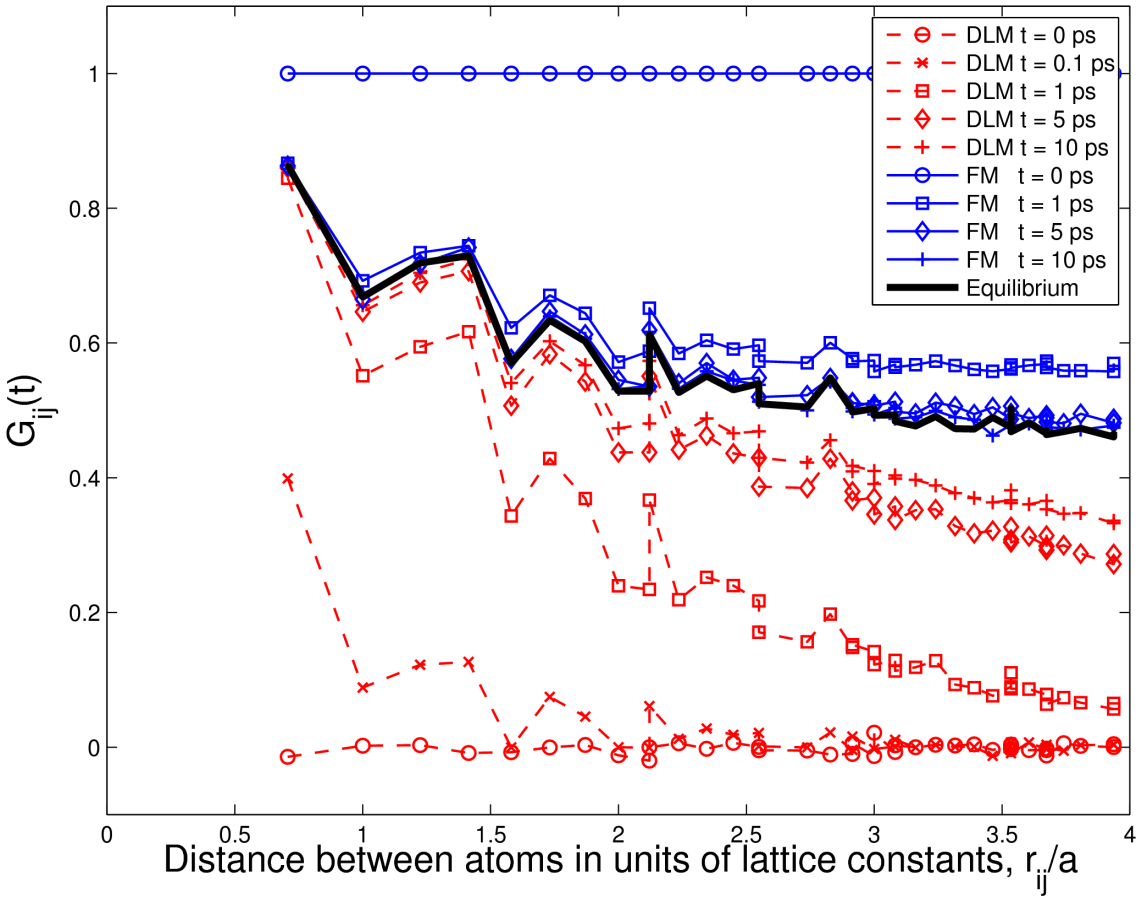
Time evolution of the equal time pair correlation function

$$G_{ij}(t) = \langle \mathbf{m}_i(t) \cdot \mathbf{m}_j(t) \rangle$$
$$= \langle \mathbf{m}(\mathbf{r}_i, t) \cdot \mathbf{m}(\mathbf{r}_j, t) \rangle$$



y=0.25%
T=100 K

below T_c

The pair correlation equilibrates faster from FM than from DLM

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Time evolution of the equal time pair correlation function

$$G_{ij}(t) = \langle \mathbf{m}_i(t) \cdot \mathbf{m}_j(t) \rangle$$
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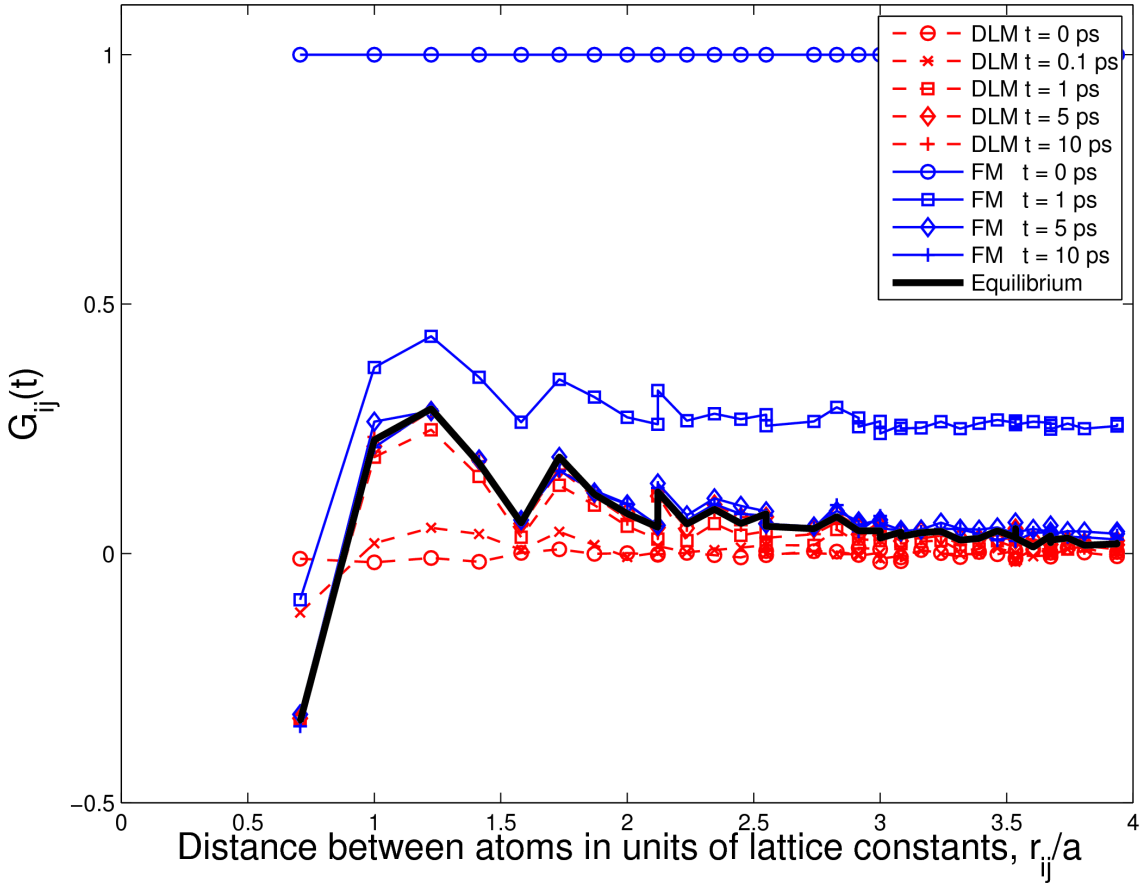

y=1.25%
T=100 K

above T_c

The pair correlation equilibrates slower from FM than from DLM

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Two-time correlation functions are powerful tools to study relaxation.

For magnetic (model Hamiltonian) systems the simplest two-time correlation is the autocorrelation

$$C_0(t_w + t, t_w) = \langle \mathbf{m}_i(t_w) \cdot \mathbf{m}_i(t_w + t) \rangle$$

The autocorrelation is not directly measurable - but relates to the zero (low field) magnetic susceptibility in experiments

$$\mathbf{M}_i(t_w, t)/h$$

The autocorrelation function has been used to study model Heisenberg Hamiltonian spin glass systems
L. Berthier, A.P. Young PRB **69,** 184423 (2004)

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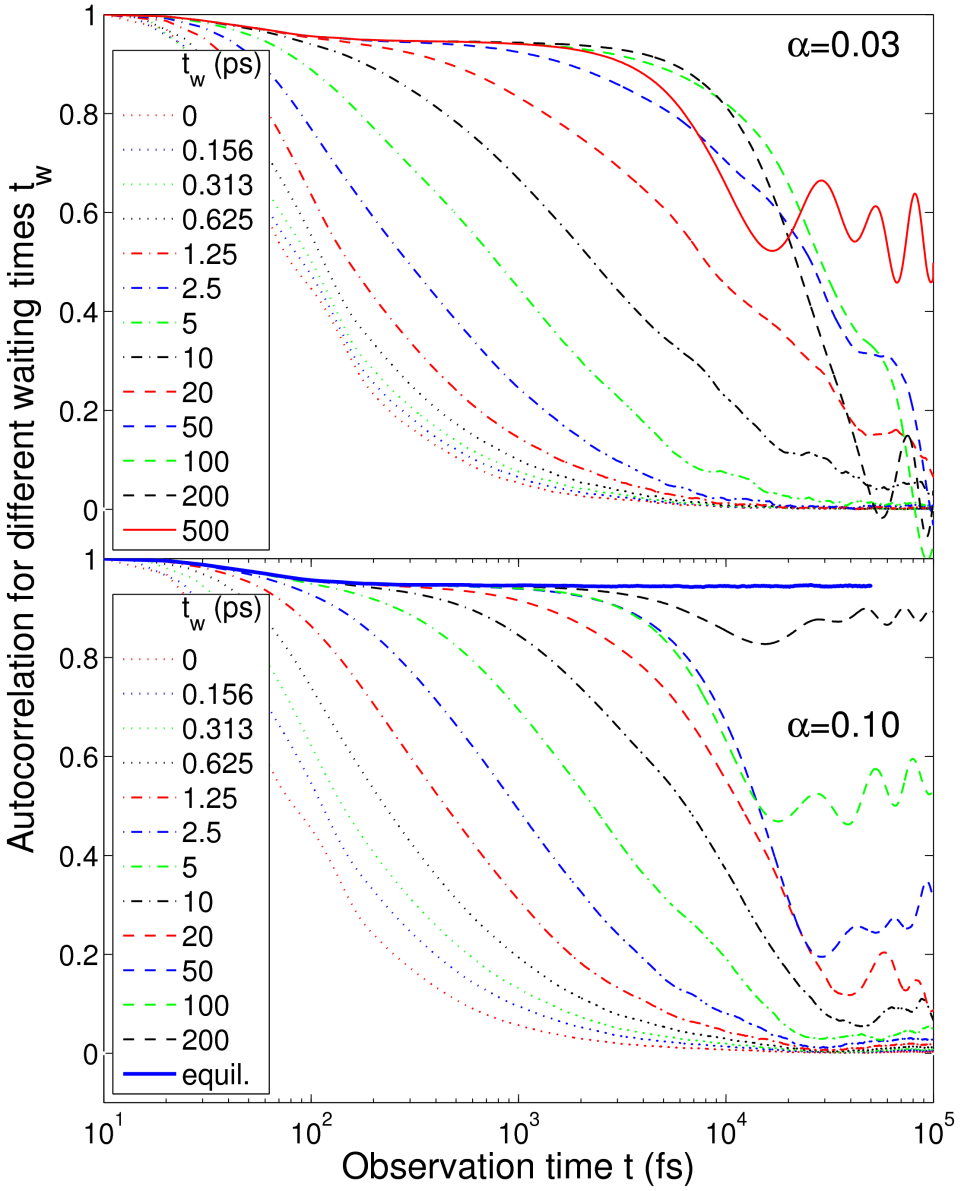
Autocorrelation C_0 y=0.25%, T=10 K ($T_c \sim 160$ K)



Quenching simulation:

Starts from DLM configuration

Waiting times logarithmically distributed

The system is ferromagnetic and relaxes similar to a simpler compound such as bcc Fe.

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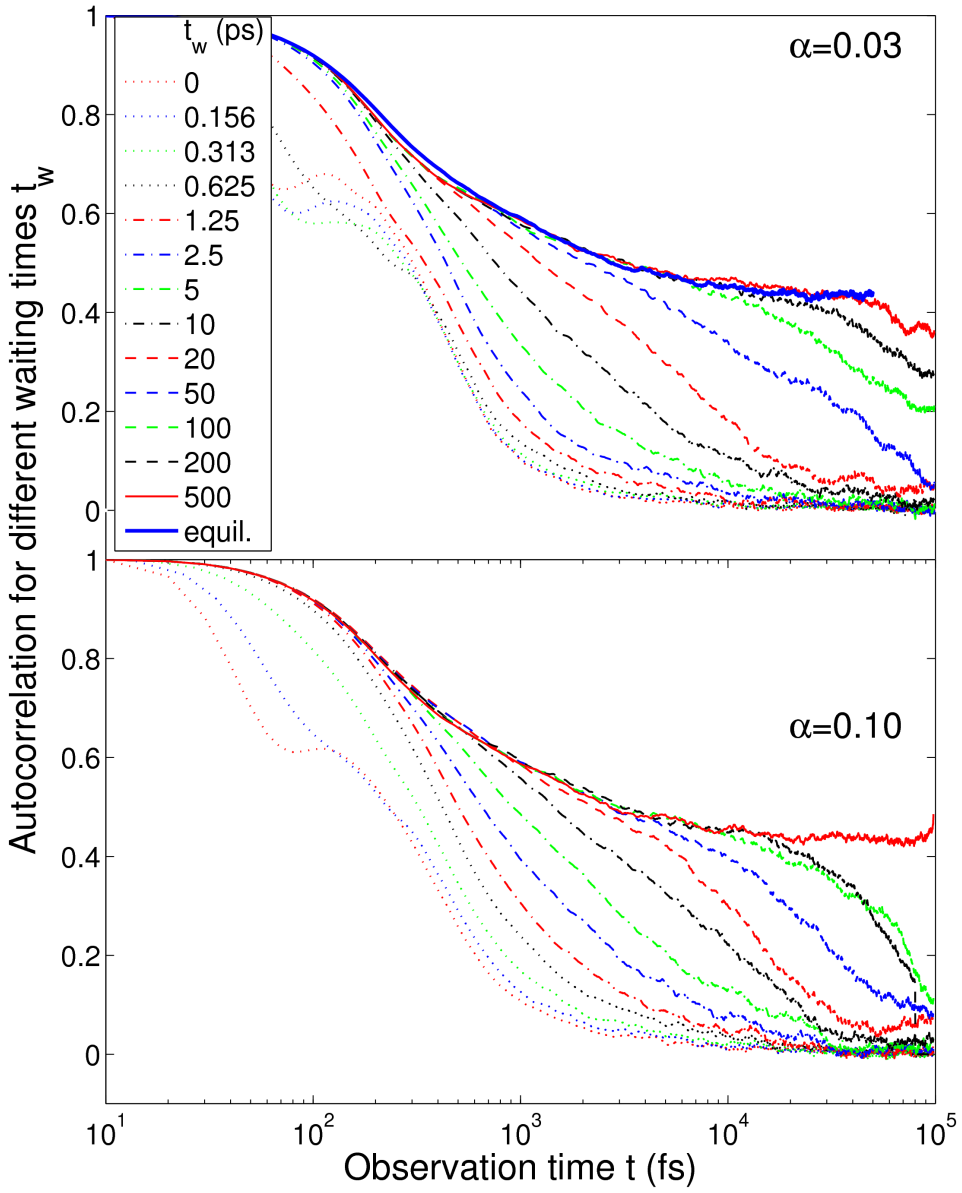
Autocorrelation C_0 y=1.75%, T=10 K (T_c~30 K)



In equilibrium the average magnetization for y=1.75% is ~0.5M_0.

This is reflected in the autocorrelation that, at large waiting times t_w, goes to the value ~0.5M_0 with increasing observation time t

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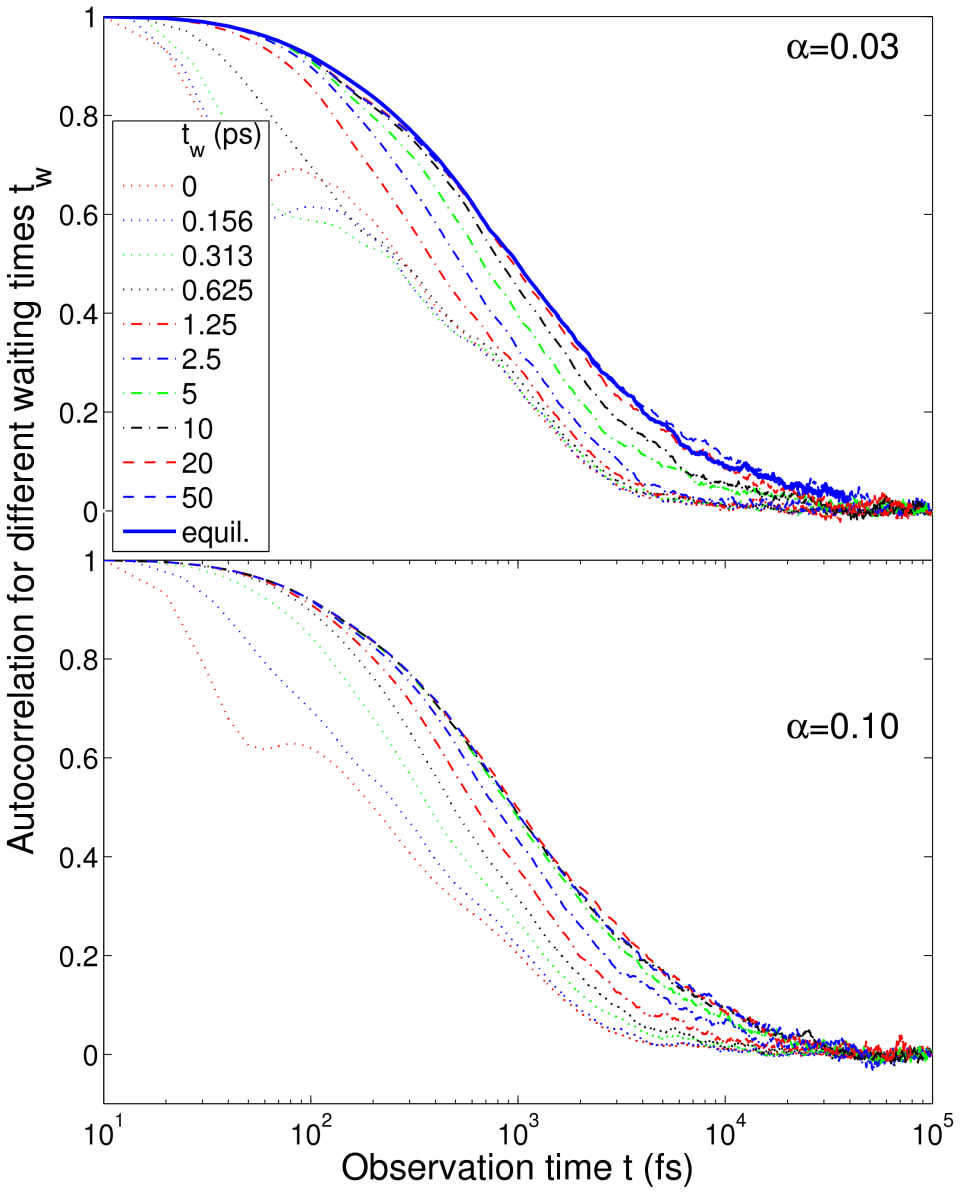
Autocorrelation C_0 y=2.00%, T=10 K (T_c<10 K)



For this antisite concentration the system is paramagnetic at T=10 K

This is reflected in the autocorrelation that, at all waiting times t_w, drops to zero with increasing observation time t

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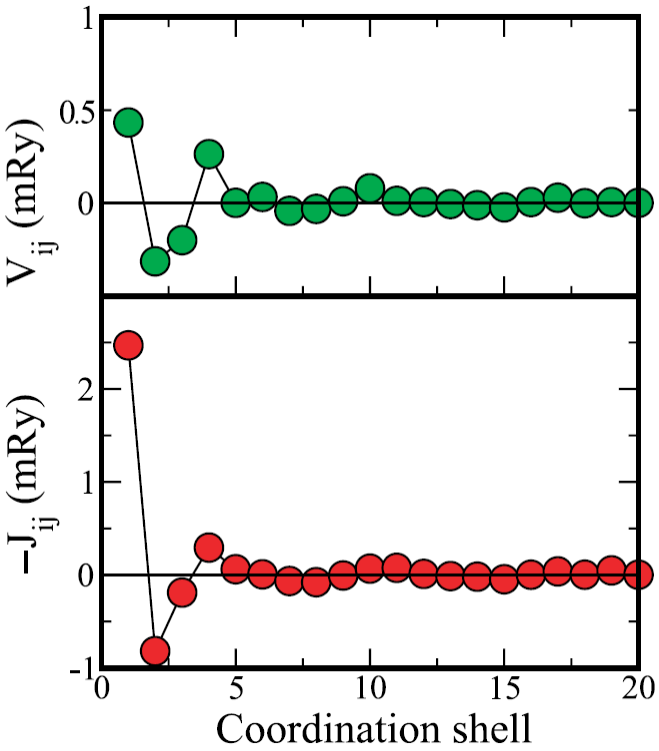
The canonical spin glass alloy CuMn:
Frustrated magnetic system without long range order

Lattice structure: fcc with Cu substituted by Mn.

Does clustering of Mn occur? Magnet short range order?



The electronic structure was investigated with EMTO

Effective pair chemical and magnetic exchange interactions were calculated.

Atomic short range order and magnetic short range order studied

Peil et al, New J. of Physics 10, 083026 (2008)

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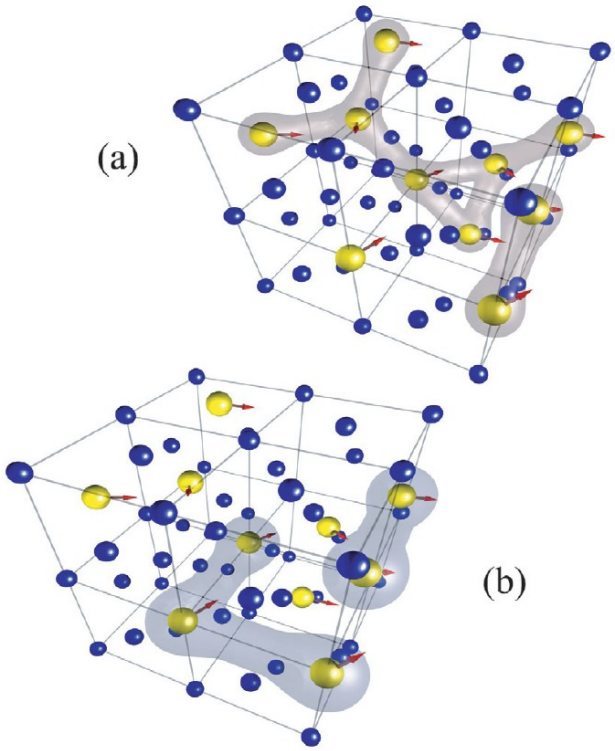
The canonical spin glass alloy CuMn

Does clustering of Mn occur? Magnet short range order?



Atomic short range order and magnetic short range order were investigated by Monte Carlo simulations.

The resulting atomic short range order shows a peak for the superstructure vector

$$\mathbf{q} = (1, 1/2, 0)$$

Magnetic short range order for the vector

$$Q_m = (1, 1/2 \pm \delta, 0)$$

Peil et al, New J. of Physics 10, 083026 (2008)

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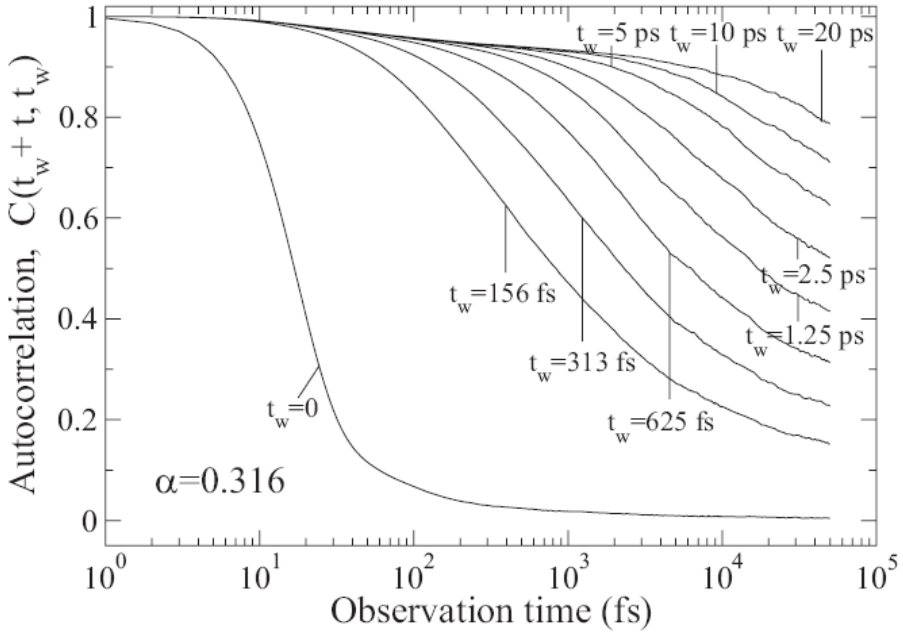


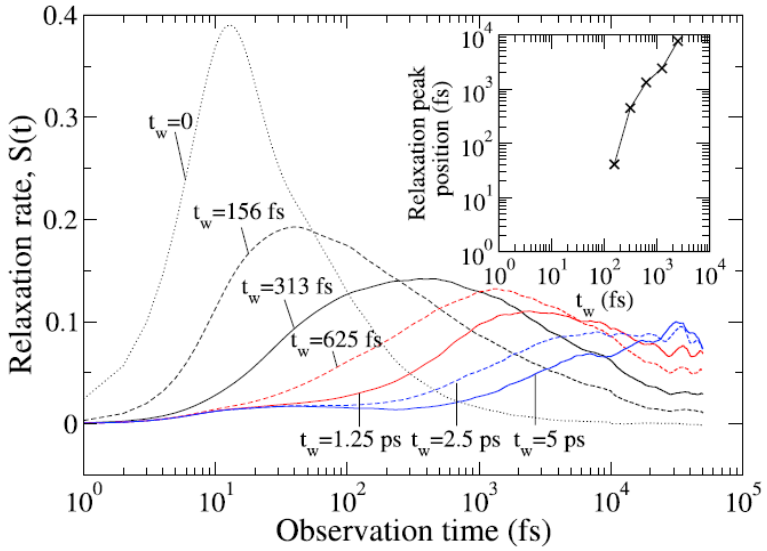
SD Simulation of the spin glass alloy CuMn

Lattice structure: fcc with Cu randomly substituted by Mn.

The autocorrelation shows slow relaxation

Define the relaxation rate function

$$S(t_w + t, t_w) = -\frac{d}{d \ln t} C(t_w + t, t_w).$$



Skubic et al., PRB **79**, 024411 (2009)

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Fitting of the autocorrelation function to two exponential functions

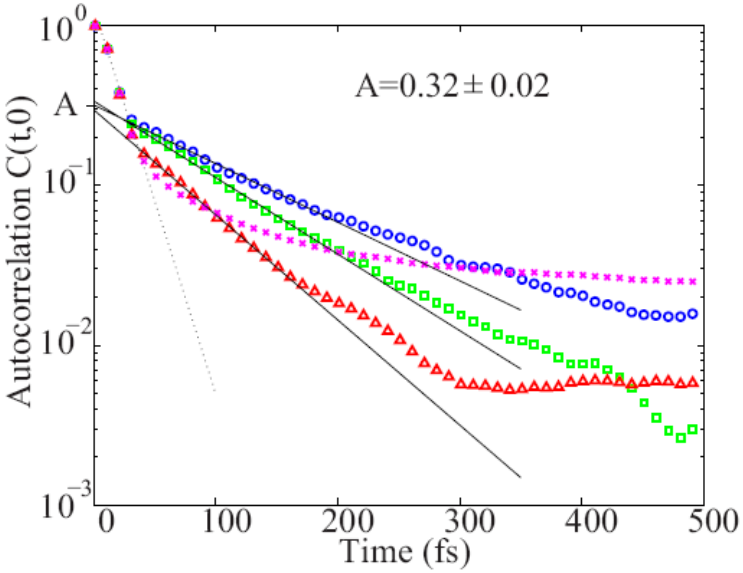

FIG. 5. (Color online) Autocorrelation $C(t, t_w=0)$ for four values of the damping parameter: $\alpha=0.01$ (circles), $\alpha=0.0316$ (boxes), $\alpha=0.1$ (triangles), and $\alpha=0.316$ (crosses). The dashed line is a linear fit to the points for 10 fs $\leq t \leq$ 40 fs; the slope is equal to $1/\tau_1$ of Eq. (12). The slope of the solid lines corresponds to the damping relaxation rate $1/\tau_2$ in Eq. (12).

$$C(t,0) \approx (1-A)e^{-t/\tau_1} + Ae^{-t/\tau_2}, \qquad (12)$$

The initial decay 0<t< 40 fs is independent of alpha and corresponds to relaxation rate $1/\text{tau}_1$

The decay 40<t<150 fs corresponds to $1/\text{tau}_2$ with tau_2 dependent on alpha

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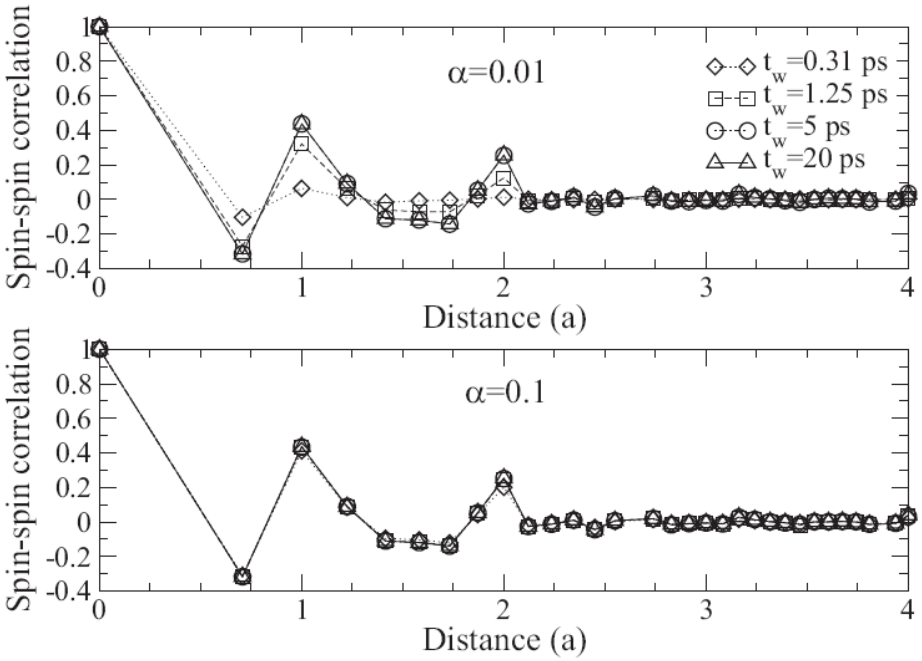
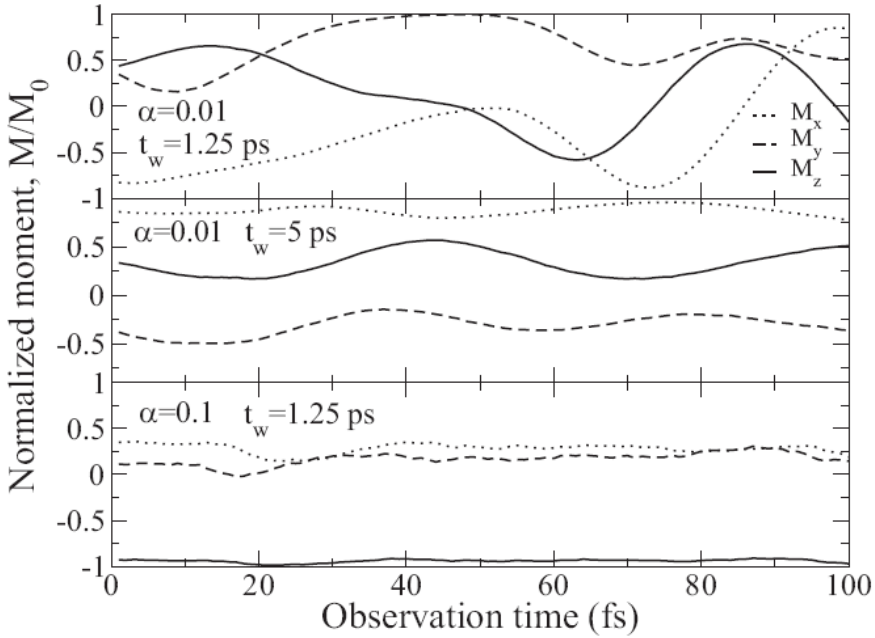
Time evolution of the spin correlation function



Typical motion of a single spin after different waiting times



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Application: Investigations on spin wave instabilities

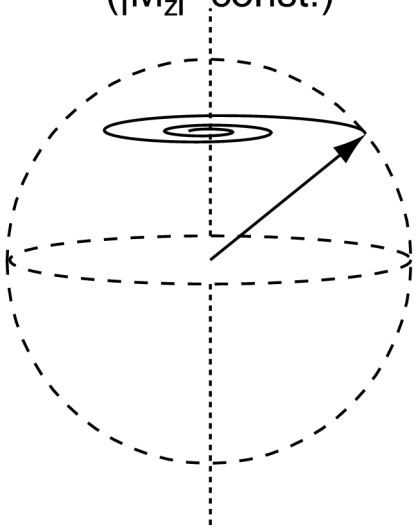
We study in simulations how the magnitude of the local magnetization can decay in precession around an anisotropy axis.

Here local magnetization is the average over the atomic magnetic moments in the simulation cell.

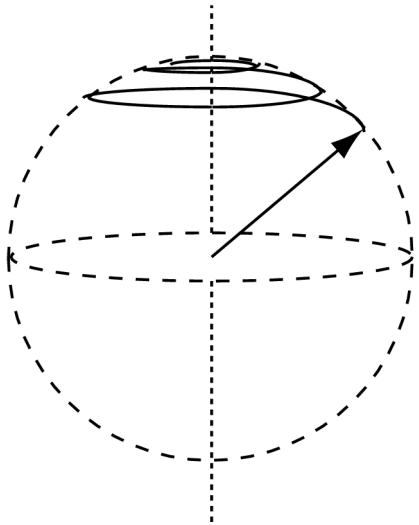
$$m_k = \frac{1}{N} \sum_i m_{k,i}$$

$$m = \sqrt{m_x^2 + m_y^2 + m_z^2}$$

a. Bloch-Bloembergen damping ($|M_z|$=const.)



b. Gilbert damping (|M|=const.)



Different Phenomeno-logical relaxation processes for the local magnetization

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These kinds of collapse of the local magnetization are referred to as a spin wave instabilities (SWI).
Interest into SWIs is motivated not least as they can enable faster switching of magnetization in applied field.

Well known underlying mechanisms for SWI are the Suhl instability, 2-magnon scattering and 4-magnon scattering. But, all these rely on the magnetostatic (dipolar) interaction!
Here we exclude dipolar interaction and the effect arise due to the magnetocrystalline anisotropy energy (MAE).

SWI caused by MAE have been studied by:
Safonov et al. **PRB** 63, 094419 (2001)
Kashuba, **PRL** 96, 047601 (2006)
Garanin et al. **EPL** 82, 17007 (2008)

In our study we focus on the behavior at finite temperature and on zero versus finite damping.

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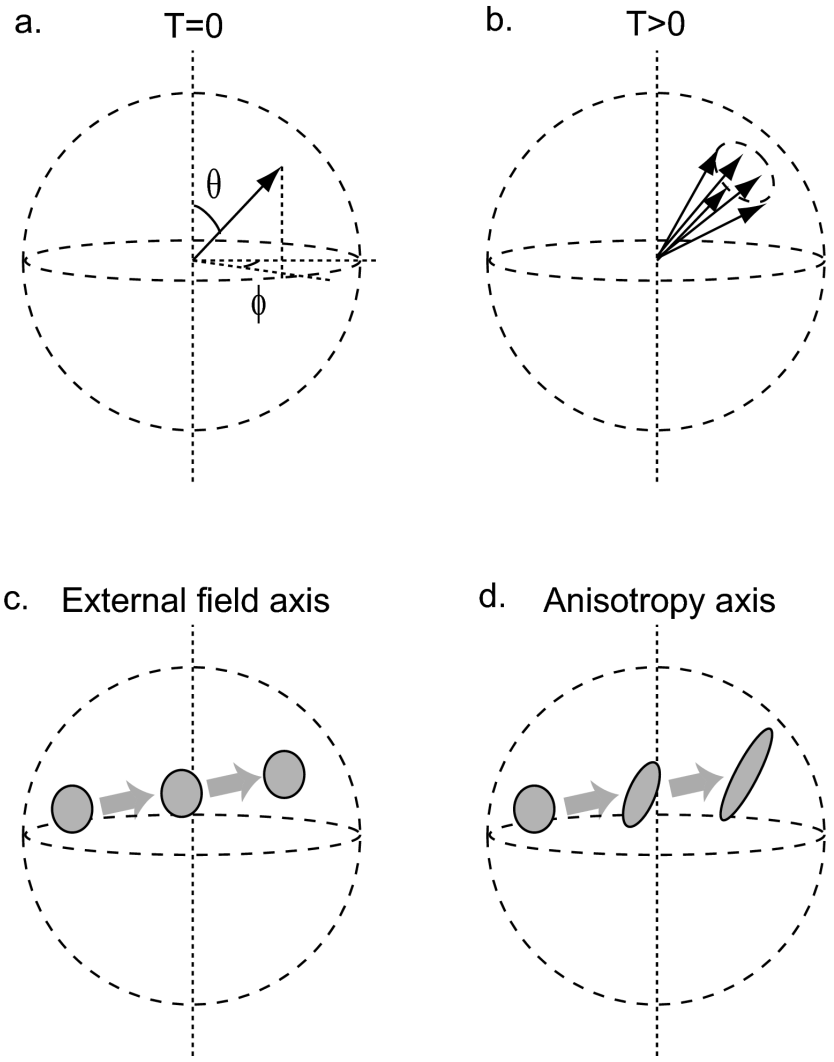

The distribution of magnetic moments at zero (a) and finite (b) temperatures.

Precession in external field (c) or around anisotropy axis (d)

The external field does not change magnetic moment distributions.

The anisotropy axis changes the moment distribution

Hellsvik et al. arXiv:0903.2186v1

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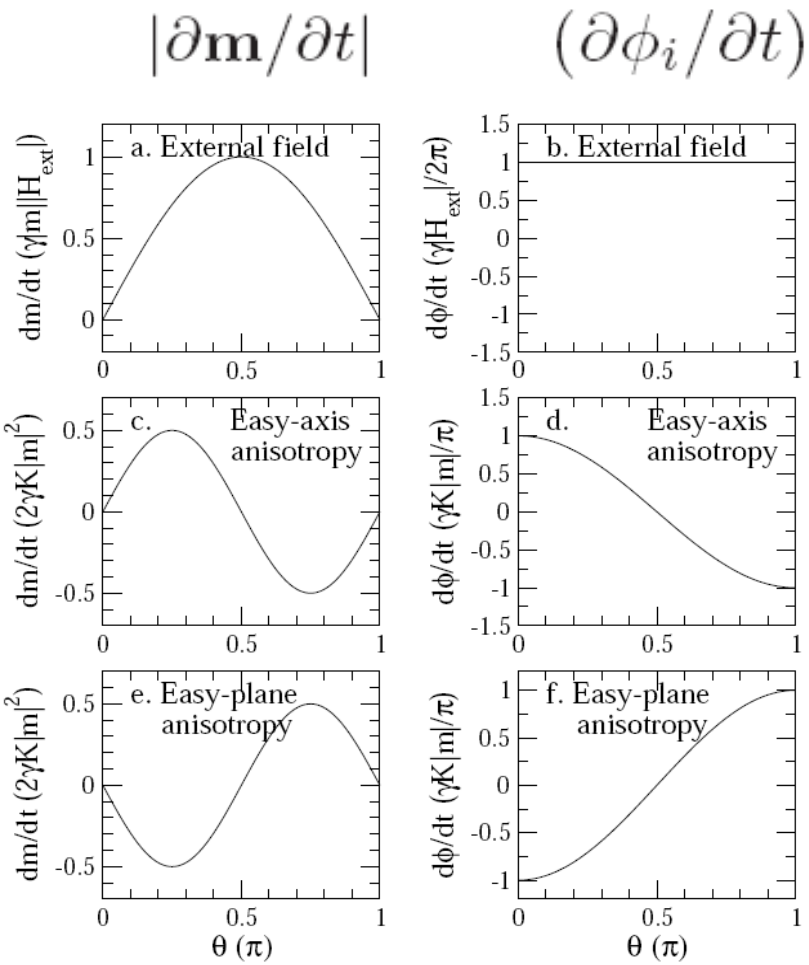

FIG. 2: The plots illustrate the change of the magnetic moment due to an external field, easy-axis anisotropy and an easy-plane anisotropy. The graphs on the left hand side give the magnitude $|\partial \mathbf{m}/\partial t|$ while the graphs on the right hand side give the angular velocity of the atomic spins with respect to angle θ between spin and applied field or anisotropy axis. Note that in the case of a uniaxial anisotropy field θ is defined as the angle between moment and a fixed crystallographic direction of the anisotropy field (e.g. 100). H is the strength of the external field and K the strength of the anisotropy field.

Change of the magnetic moment due to:
External field
Easy axis anisotropy
Hard axis anisotropy

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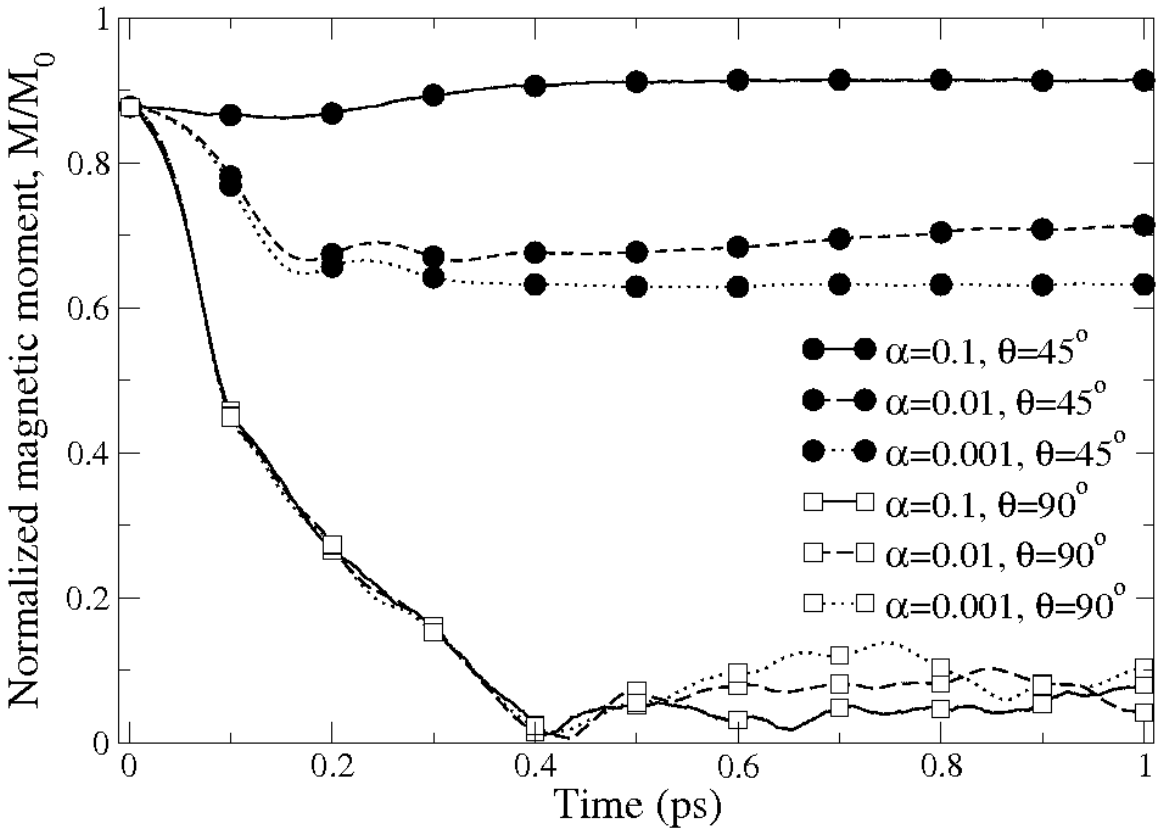
Relaxation of magnetization starting with the local magnetization at angle 45 or 90 degrees to the uniaxial easy anisotropy axis.

These simulations for an unrealistically high value of the MAE, Ku=-2.0mRy/atom (xxx MJ/m³)



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These simulations with 10 times smaller MAE

Also here the value of the damping is crucial

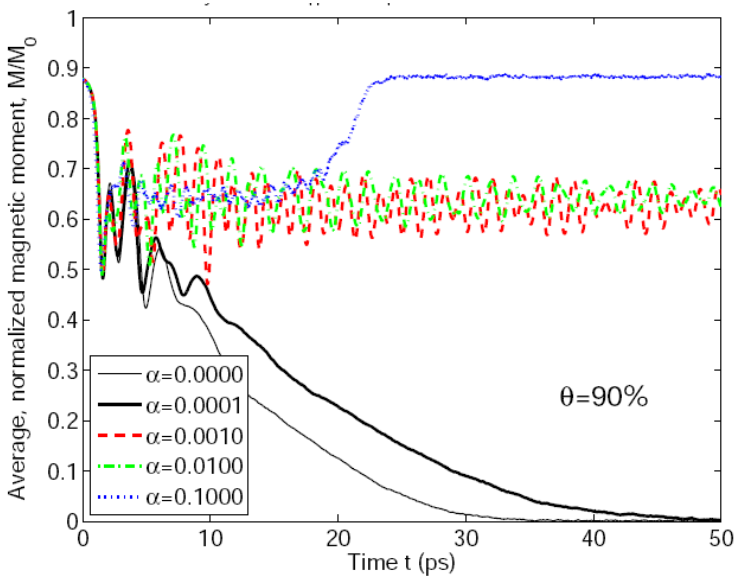

Figure 15: Uniaxial anisotropy -0.2 mRy, $\theta = 90°$, $\Delta t = 10$ as.

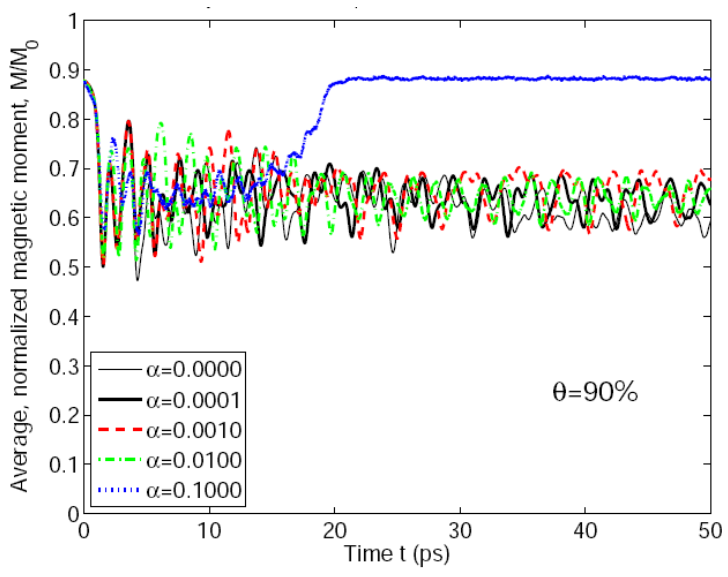

Figure 16: Uniaxial anisotropy -0.2 mRy, $\theta = 90°$, $\Delta t = 1$ as.

Dependence on time step used in SDE solver!

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Dissipation of magnetic energy?

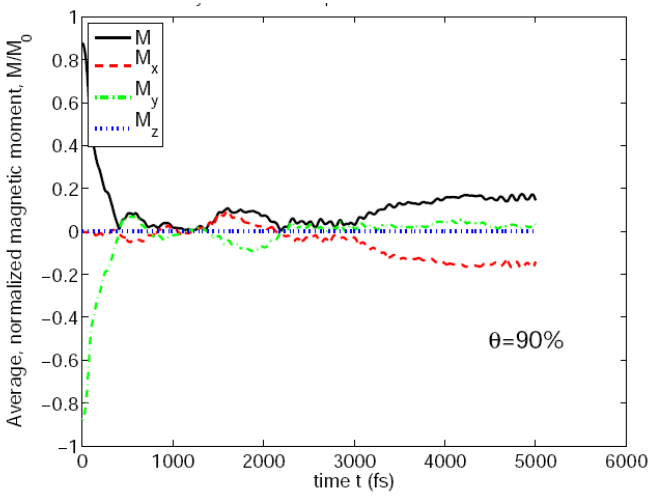

Figure 19: Uniaxial anisotropy -2.0 mRy, $\theta = 90°$, $\Delta t = 1$ as, $\alpha = 0$.

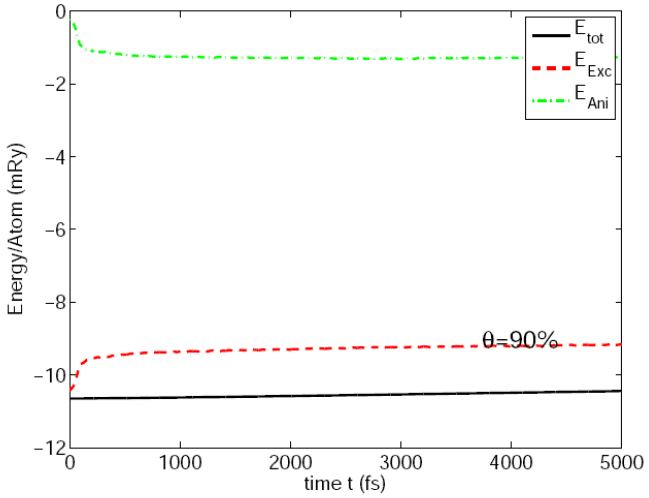

Figure 20: Uniaxial anisotropy -2.0 mRy, $\theta = 90°$, $\Delta t = 1$ as, $\alpha = 0$.

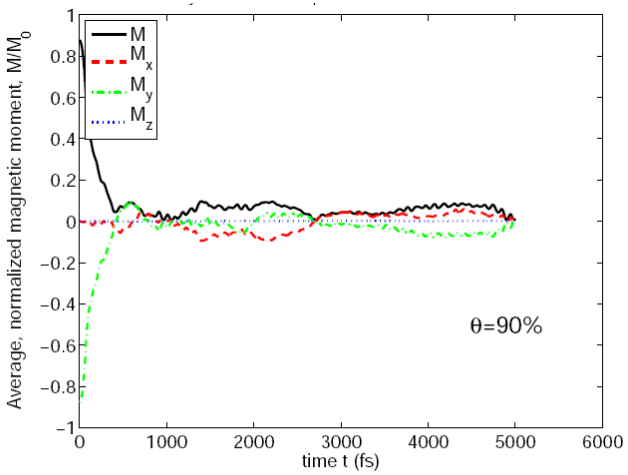

Figure 21: Uniaxial anisotropy -2.0 mRy, $\theta = 90°$, $\Delta t = 1$ as, $\alpha = 0.0001$.

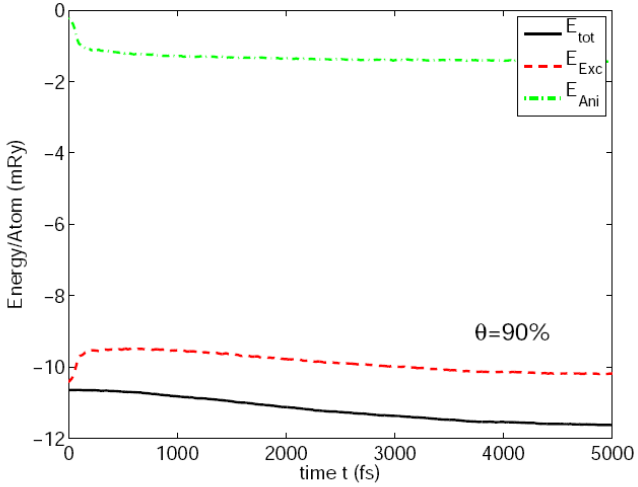

Figure 22: Uniaxial anisotropy -2.0 mRy, $\theta = 90°$, $\Delta t = 1$ as, $\alpha = 0.0001$.

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Conclusions

- Implementation of an atomistic spin dynamics, at finite temperatures, method
- Application to the dilute magnetic semiconductor Mn-doped GaAs corresponds to Monte Carlo results – and yields additional information
- The slow magnetic relaxation of the alloy CuMn reveals its character as a spin glass compound.
- Spin wave instabilities in presence of magnetocrystalline anisotropy depends on geometry, MAE strength and the damping.
- In simulations so far, complete shrinking of the macro spin occur only for unrealistically high values of the MAE.. But, incomplete SWI phenomena present also for more realistic values of the MAE.

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Outlook

- Integration with real-space electronic code for calculations on finite (very small) systems.
- Inclusion of bi-quadratic and higher order terms in Heisenberg parametrization within generalized perturbation method.
- Sharing of code – User manual and binaries posted on

  http://www.fysik.uu.se/cmt/webserver4/index.php

  Source code available upon request.

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Numerical simulations have been performed on computer systems at UPPMAX and NSC.