Simulation of self-assembly of superparamagnetic particles

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Supported by DOE grant No. DE-FG02-98ER45714 and NSF-MRSEC DMR-9809423
Outline

• Motivation: high density magnetic storage using nm FePt particles
  • Effects of superparamagnetism on self-assembly kinetics: chaining, chain stacking

• Problem: disparity of time scales (magnetization fast, motion slow)

• A solution: time coarse-graining
  • In slowly varying field, treat precession exactly
  • Arrhenius law for reversals comes out without *ad hoc* assumptions
  • Allows simulating long enough to see self-assembly
Motivation


• Various structures: hexagonal monolayers, multilayers, square arrays.

• What is the role of superparamagnetic fluctuations in self-assembly?

• Need to simulate over very long time scales compared to magnetic dynamics

FeCoPt particle arrays on TEM grids, courtesy M. Chen
Physics needed for self-assembly simulation of superparamagnetic particles

• Magnetization dynamics
  • Landau-Lifshitz equation for $dM/dt$
  • $H = $ magnetostatic + external + thermal noise

• Particle motion
  • Magnetic torque $\mathbf{M} \times \mathbf{H}$, force $q\mathbf{H}$ (2 monopoles $q$)
  • Steric force: hard core
  • Hydrodynamic force
  • Random Langevin force
Problems of disparate time scales

Short time scale: magnetic precession time (ns)

Long time scale: particle motion, self-assembly (s)

Solution of precession-time problem: we need to allow \( \Delta t \gg \text{precession period} \) \( (\gamma H \Delta t \gg 1) \).

That requires us to “average out precession”.
Averaging out precession

1. Arrhenius-Neél approach?
   - Assumes \( \mathbf{M} \) lies at minimum of Stoner-Wohlfarth energy
   - \( \mathbf{M} \) jumps between minima with probability \( \omega \exp(-\Delta E/k_B T) \)
     where \( \Delta E(\mathbf{H}, \mathbf{M}) \) is the energy barrier and \( \omega \) is an “attempt frequency”.
   - Theoretical problems: what is \( \omega \)? For isotropic particles, what are minima?
   - Practical problem: such a jump sends a discontinuous shock through the system that is difficult to deal with numerically.
Averaging out precession


Evolve the system exactly over interval $\Delta t$.

Effectively averages out fast precession, but still treats slow precession correctly. Use this.
Time coarse-graining

Starts from the Landau-Lifshitz equation:

\[
\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H} - \frac{\gamma\alpha}{M_s} \mathbf{M} \times (\mathbf{M} \times \mathbf{H})
\]

Ignores rapidly varying parts of the local field \( H \) (but correctly describes rapid precession of \( M \)).

Uses Kikuchi’s exact solution (for constant \( H \)) to the LL equation:

\[
\alpha \gamma H t = \frac{1 - \cos \theta(t)}{1 + \cos \theta(t)} \quad \mathbf{M}_z = M_s \cos \theta(t)
\]

\[
\alpha \gamma H t = \frac{\mathbf{M}_x - i\mathbf{M}_y = M_s \sin \theta(t) e^{i\gamma H t}}{1 + \cos \theta(t)}
\]

From \( \mathbf{M}, \theta \) at time \( t \), it is easy to calculate them at \( t + \Delta t \) even if \( \gamma H \Delta t \) is large.

What is being left out: resonant energy transfer (small). \( \alpha = 0.1 \)
Magnetization averaging

The effects on the other particles are determined by the average magnetic moment

\[ \overline{M_x} - i\overline{M_y} = M_s \int_{t}^{t+\Delta t} \sin \theta(t) e^{i\gamma H t} \]

which cannot be analytically integrated, but there is a simple closed-form parameterization that can (error < 1%):
Large-\(\Delta t\) simulation

Advantages of this time coarse-graining scheme:

- exact at small \(\Delta t\)
  \((\gamma H \Delta t = 0.0314\) movie\)
- exact at large \(\Delta t\)
  \((\gamma H \Delta t = 31.4\) movie\)

Damping \(\alpha = 0.1\)

(click on a sphere for movie, or see http://bama.ua.edu/~visscher/colloids)

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Particle Interactions

M’s precess around each other, eventually becoming parallel (lowest energy configuration, attractive interaction)

$\alpha = 0.001$ movie

$\alpha = 1.0$ movie
Finite temperature: add Langevin noise to LL equation

Start from the minimum-energy configuration (parallel M’s).

Movie shows energy building up until it’s taken away by $\alpha$ as fast as it’s created by thermal noise.
Self-assembly!

Put 50 particles in a box, with one (Lennard-Jones) attractive wall. Movie begins with random particle positions.
Self-assembly!

Final frame of movie: nearly perfect hexagonal lattice (top view).

For this case (strongly superparamagnetic, large $a$ hence short dephasing time) the magnetic interactions tend to average out – get hexagonal lattice.
Summary

• We have developed an algorithm for following superparamagnetic dynamics over the long time scales of self-assembly, without following the precession in detail.

• This is done by using an exact constant-field solution to evolve the system over a time step that can be long compared to the precession period.

• The resulting scheme can successfully model the self-assembly of superparamagnetic particles.