# Role of the dipolar interaction in magnetic hyperthermia

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Motivation

◆ I want to tell you about a model to describe the dyamics of *small* magnetic particles at *finite temperature*.

   ◆ For small particles, temperature makes *all* the difference.

◆ This model can simulate any external condition. So we can describe any experiment:

   ◆ Magnetometry, FMR, ZFC-FC, AC Suscep, Hyperthermia, &c.

◆ Having a good model is important for fundamental reasons:

   ◆ We need to understand how temperature affects the magnetic properties.

◆ ... and is also of applied interest:

   ◆ How to build better materials for hyperthermia.
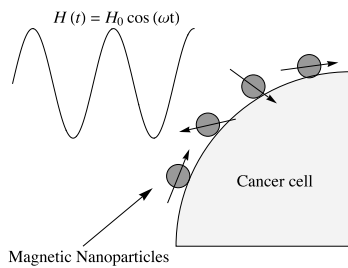
Exchange vs. Magnetostatic

◆In any magnetic material there is a competition between the exchange energy and the magnetostatic energy.

Exchange energy

◆The exchange energy stems from the overlap of the electronic wave functions. The energy between two spins S_1 and S_2 is

$$E_{\text{exch}} = -J\, S_1 \cdot S_2$$

◆This energy favours the parallel alignment of the spins.

Magnetostatic energy

◆The magnetization of the material creates a field which interacts with itself.

◆This is the long-range dipolar interaction that acts on the system.

```
(*load["DipolarCartoon.pdf",0.3]*)
Show[

 StreamPlot[{ (3 x z)/(x² + z²)^(5/2) , (3 z²)/(x² + z²)^(5/2) - 1/(x² + z²)^(3/2) }, {x, -2, 2}, {z, -1, 1},

   StreamStyle → Gray, Frame → False,
   AspectRatio → 1 / 2
 ],
 Graphics[{
    Lighter@Orange, Disk[{0, 0}, {0.2, 0.35}],
    Black, Arrowheads[0.022], Arrow[{{0, -0.14}, {0, 0.18}}]

 }],
 ImageSize → 650,
 PlotLabel → Style["Anti-parallel configuration", 24]
]
```
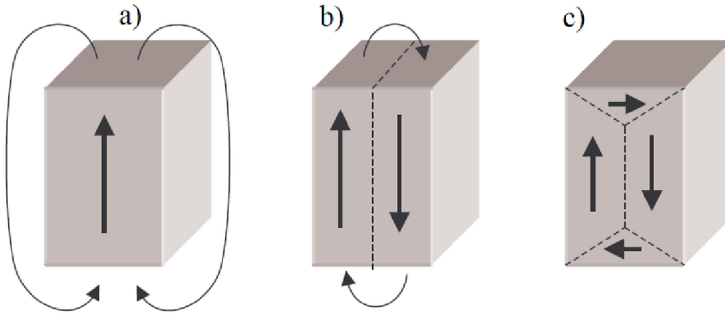
Anti−parallel configuration
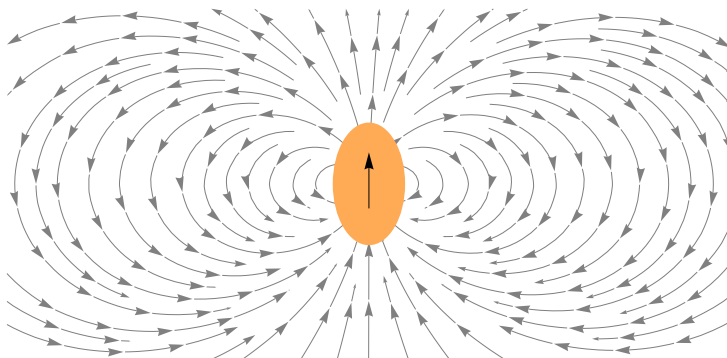
◆ This energy favours a disordered configuration.

Fight!

◆ These two energies compete.
◆ This may lead to the formation of *magnetic domains*.

What if the object is small?

◆ If the object is small the **exchange energy wins**.
◆ The system behaves as a *single magnetic domain*.
  ◆ $10^3 - 10^4$ atoms acting in *unison* (I like this word!).

Magnetization

◆ In general the magnetization of a material is a function of r and t: $M(r, t)$.
◆ But for a single-domain the magnetization is uniform inside the particle.
◆ Thus we are left with a single vector $M(t)$.
◆ Define

$$m(t) = \frac{M(t)}{M_S}$$

◆ Them $m(t)$ is a unit vector. The dynamics take place in the *unit sphere*.

Anisotropies

- The exchange interaction is *isotropic*. It says
  - "We spins stay aligned! But I don't care in which direction."
- But that is not what we observe experimentally.
- What we see is that there are certain preferred directions for the magnetization to lie on.
- This is due to the presence of *anisotropies*.

Crystal field and shape

- There are usually two contributions to the anisotropy.
  - The crystal lattice may have preferred orientations that favour the alignment of ***m*** in certain directions.
  - The shape of the particle may have the same effect (e.g. ellipsoidal particles).

Uniaxial Anisotropy

- The most common case, and the one we will discuss here, is that of uniaxial anisotropy.
  - One axis has a preferred direction.
  - We define it as the ***z* axis**.
- It doesn't matter if ***m*** is pointing at $+z$ or $-z$.
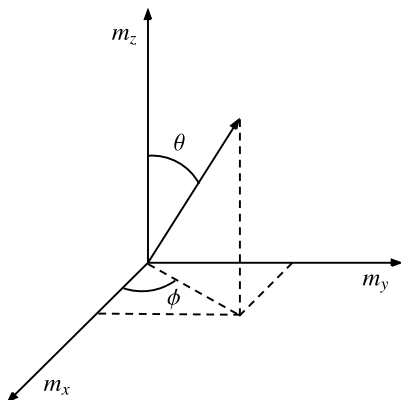- We usually write the energy as

$$E_{\text{uni}} = -K \, V \, m_z^2 \; = \; -K \, V \cos^2 \theta$$

where

$$K \;=\; \text{Anisotropy constant} \left(\text{units of } J/m^3, \; \text{erg}/\text{cm}^3, \; \text{etc.}\right)$$
$$V \;=\; \text{Volume of the particle}$$

- This energy has minima at $m_z = \pm 1$ or $\theta = 0, \pi$.

- If we also have an externally applied magnetic field ***H***$_a$ then

$$E = -V \, \boldsymbol{M} \cdot \boldsymbol{H}_a - K \, V \, m_z^2$$

Superparamagnetism (thermal instability)

◆ The uniaxial anisotropy has 2 minima.
◆ The energy barrier that sepparate them has height KV.
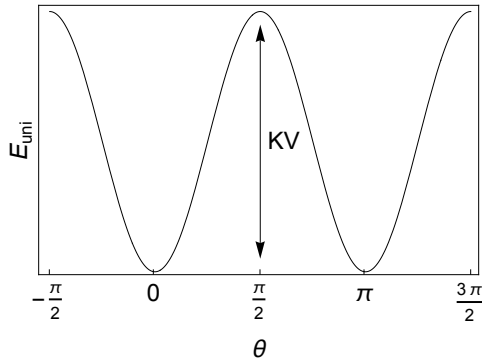
Anisotropy barrier vs. thermal energy

◆ Define the ratio between the anisotropy barrier and the thermal energy:

$$\sigma = \frac{KV}{k_B\,T}$$

◆ When V is small → σ **may be small**.

```
Row[{

Animate[
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     FrameTicks → {{-π/2, 0, π/2, π, 3 π/2}, None, None, None},
     PlotRange → {-1.1, 0.1},
     PlotLabel → "Low barrier (or Hot)",
     Epilog → {
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       }
     ],
   {i, 1, Length[OW1[[1]]], 1},
   AnimationRunning → False,
   DefaultDuration → 200
   ],
  Spacer[100],
  Animate[
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     FrameLabel → {"θ", "E_uni"},
     FrameTicks → {{-π/2, 0, π/2, π, 3 π/2}, None, None, None},
     PlotRange → {-1.1, 0.1},
     PlotLabel → "High barrier (or Cold)",
     Epilog → {
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       }
     ],
   {i, 1, Length[OW2[[1]]], 1},
   AnimationRunning → False,
   DefaultDuration → 200
   ]}]
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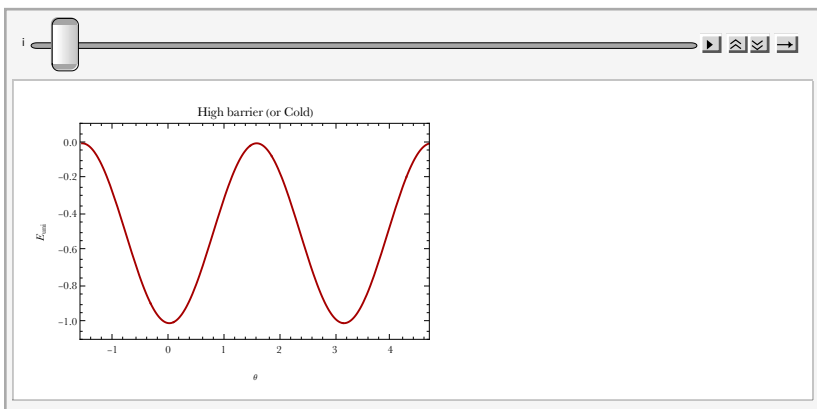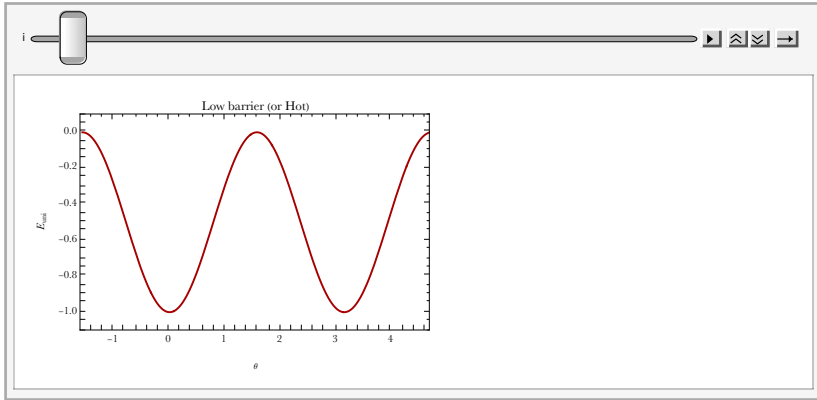
Superparamagnetism

- This is the phenomena of *superparamagnetism*.
- There is *no* stable magnetic configuration.
- Due to the aggitation, if you wait long enough you can always observe a *spin flip*.
- Of course, depending on the material it may take 10 million years
  - This is what we expect of a *permanent magnet*!

Two possibilities

1. Brown relaxation: the magnetization vector is held fixed, and the particle physically rotates

2. Néel relaxation: the particle physically does not move; only the magnetization vector rotates

- Both occur in real systems
- But depending on the size one or the other is predominant.
- In this talk I will discuss only Néel relaxation.
- It is the most important for hyperthermia.

Dynamics

3 steps: naive → naive + damping → naive + damping + flucutuations

Round 1: naive approach

♦ Big question: how to describe the dynamics of $m(t)$ under the influence of temperature?
♦ Since all spins operate in unison

$$m(t) \propto \sum \langle S_i \rangle$$
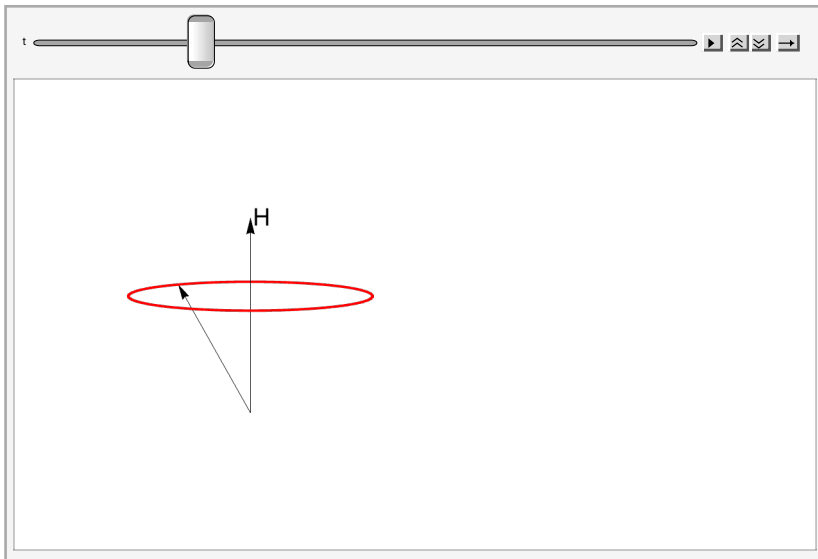
♦ Hence from the Heisenberg equation,

$$\frac{dm}{dt} = -\gamma \, m \times H$$

where

$$\gamma = 2.2 \times 10^5 \, \frac{m}{As}$$

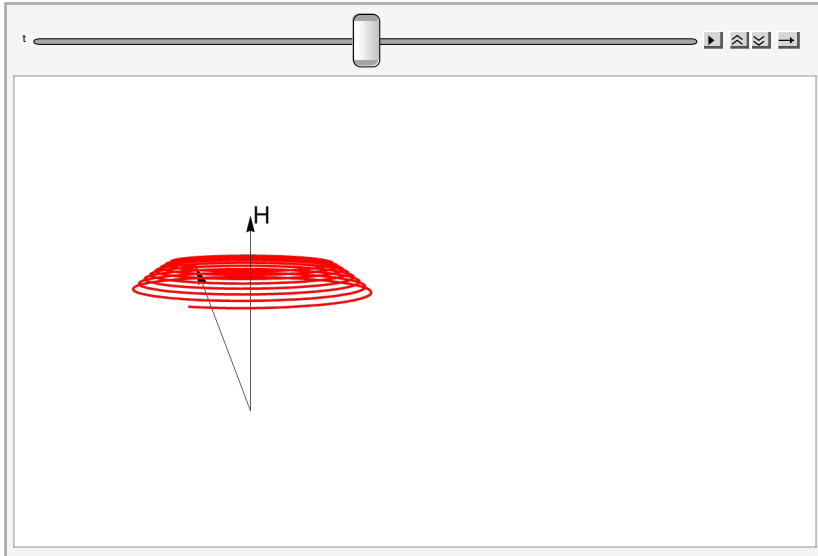is the gyromagnetic ratio of the electron.
♦ This equation is non-dissipative.
♦ It predicts the *precession* of the magnetization around **H**.
♦ It preserves $|m| = 1$

Round 2: damping

♦ We know the magnetization does not precess indefinitely.
♦ There is a *torque* that makes **m** align with **H**.
♦ So if we start the system in a given point, it will eventually stop.
♦ There is *damping*.

◆ The physical origins of the damping is still an open discussion.
  ◆ Spin interacts with stuff around it: conduction electrons, phonons, nuclear spins, &c.
  ◆ This interaction is usually mediated by the spin-orbit interaction.
◆ This damping can only be introduced in the dynamical equation *phenomenologically*.
  ◆ This is the *Landau-Lifshitz* equation

$$\frac{d\boldsymbol{m}}{dt} = -\gamma\,\boldsymbol{m}\times\boldsymbol{H} - \gamma\alpha\,\boldsymbol{m}\times(\boldsymbol{m}\times\boldsymbol{H})$$

◆ Here $\boldsymbol{H}$ means the *effective field = external + anisotropy*

$$\boldsymbol{H} = -\frac{1}{V}\frac{\partial E}{\partial \boldsymbol{M}} = \boldsymbol{H}_a + \boldsymbol{H}_{\mathrm{ani}}\, m_z\, \boldsymbol{e}_z$$

where $\boldsymbol{H}_{\mathrm{ani}}$ is the *anisotropy field*.

$$\boldsymbol{H}_{\mathrm{ani}} = \frac{2\,K}{M_S}$$

◆ This equation also preserves $|\boldsymbol{m}| = 1$. That is something we always want to be true.
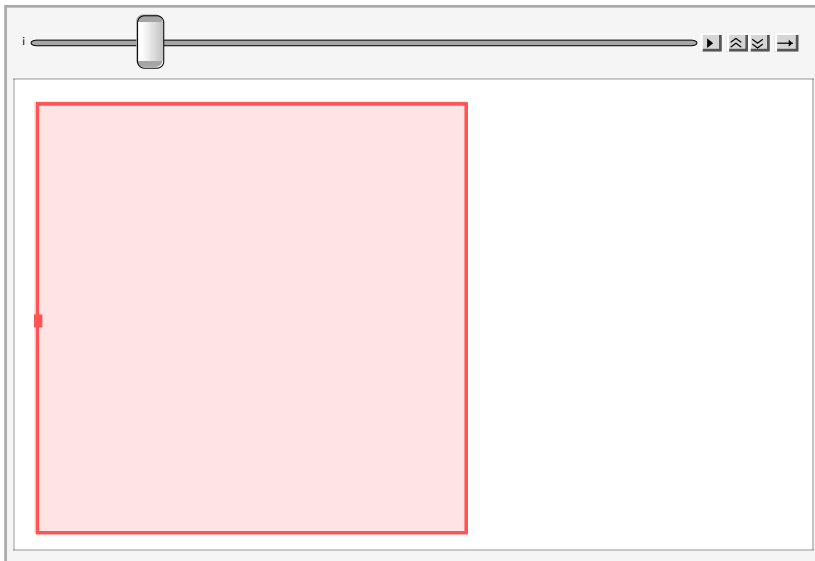
Round 3: thermal fuctuations

Before I talk about it, let us look at an analogy

Brownian motion and Brown's theory

Two Browns.

Robert Brown (1827)

◆ Observed the "rapidly oscillatory motion" of grains of pollen in water.

◆ Langevin introduced a great model. His reasoning was:
  ◆ Pollen is classical! Newton's law must hold!
  ◆ But we are missing a **force**!
  ◆ This extra force must be random.
  ◆ It is present in all systems, but only becomes significant if the system is small.
◆ In Newtonian mechanics:

$$\text{Round 1}: \frac{dp}{dt} = F$$

$$\text{Round 2}: \frac{dp}{dt} = F - \alpha p$$

$$\text{Round 3}: \frac{dp}{dt} = F - \alpha p + \xi(t)$$

◆ We add a **random force** $\xi(t)$.
  ◆ It describes the many collisions of the grain of pollen with the molecules in the medium.
◆ $\xi(t)$ must be insensitive to the origin of time so

$$\langle \xi(t) \rangle = 0$$

◆ Moreover we assume that $\xi(t)$ and $\xi(t + \Delta t)$ are uncorrelated: there are many many molecules!

◆ Finally, we assume that $\xi(t)$ is drawn from a Gaussian distribution. This is based on the *central limit theorem* which says that:

> *"When random things get complicated, they become Gaussian."*

  ◆ This is why the errors in the physics lab are usually Gaussian.
◆ But since $\xi(t)$ appears in a differential equation, it must be infinite in order to give a finite contribution.
◆ Everything is summarized in

$$\langle \xi(t)\,\xi(t')\rangle \;=\; 2\,D\,\delta(t-t')$$

where D is called the *diffusion constant.*

◆ Langevin was able to find D by making a connection with thermal equilibrium: after a long time has passed, the momentum distribution must be given by Gibbs formula (the Maxwell-Boltzmann distribution). The result is:

$$D \;=\; \gamma\,m\,k_B\,T$$

Some points worth noting

◆ The thermal fluctuations have the same origin as the bath:
  ◆ The interaction with the environment.
◆ Ideally we could try to describe all particles. But that is too complicated.
◆ So we "integrate" over the bath; that is, we reduce the effect of the bath to a handful of parameters.

William Fuller Brown, Jr. (1963)

◆ He is the "father" of micromagnetism. When you use OOMMF, thank him for it.
◆ He saw the Landau-Lifshitz equation and made the connection with Brownian motion.
◆ It maybe went somewhat like this:
  ◆ "Hey, I need to add a random force to the LL equation!"
  ◆ "But wait, I can't: a random force would *not* preserve $|\boldsymbol{m}| = 1$."
  ◆ "Then I must add a random field!"
◆ We then arrive at the *Stochastic Landau-Lifshitz* (SLL) equation

$$\frac{d\boldsymbol{m}}{dt} = -\gamma\,\boldsymbol{m}\times[\boldsymbol{H}+\eta(t)] - \frac{\gamma\alpha}{M_S}\,\boldsymbol{m}\times\{\boldsymbol{m}\times[\boldsymbol{H}+\eta(t)]\}$$

◆ This *random field* $\boldsymbol{\eta}(t)$ must behave just like in the Langevin theory:

$$\langle \eta_i(t)\rangle \;=\; 0$$

$$\langle \eta_i(t)\,\eta_j(t')\rangle = \frac{2\,\alpha\,k_B\,T}{\gamma\,M_S\,V}\,\delta_{ij}\,\delta(t-t')$$

◆ The constant appearing here was determined by Brown, again assuming that in equilibrium the distribution must be that of Gibbs.

Stochastic Landau-Lifshitz (SLL) equation

Success!

◆ We now have a complete model to describe the magnetization based only on fundamental constants that can be measured experimentally.

◆ The SLL is a stochastic equation: every time you simulated it, you get a different answer.

　◆ This makes sense since the thermal agitation is random.

Reduced variables

◆ We are already working with $\boldsymbol{m} = \frac{\boldsymbol{M}}{M_S}$.

◆ It is also convenient to work with

$$\boldsymbol{h} = \frac{\boldsymbol{H}}{M_S}$$

◆ We also use a reduced unit for time:

$$\tau_0 = \frac{1}{\alpha \gamma M_S}$$

◆ Then the SLL becomes

$$\tau_0 \frac{d\boldsymbol{m}}{dt} = -\frac{1}{\alpha} \boldsymbol{m} \times [\boldsymbol{h}(t) + \eta(t)] - \boldsymbol{m} \times \{\boldsymbol{m} \times [\boldsymbol{h}(t) + \eta(t)]\}$$

$$\boldsymbol{h}(t) = \boldsymbol{h}_a(t) + \lambda\, m_z, \qquad \lambda = H_{\mathrm{ani}} / M_s = 2\,K / M_s^2$$

◆ And the new $\eta(t)$ becomes

$$\langle \eta_i(t) \rangle = 0$$
$$\langle \eta_i(t)\, \eta_j(t') \rangle = 2\,\alpha\,\theta\,\delta_{ij}\,\delta(t - t')$$
$$\theta = \frac{k_B\,T}{M_S^2\,V}$$

◆ So now we have the following list of parameters

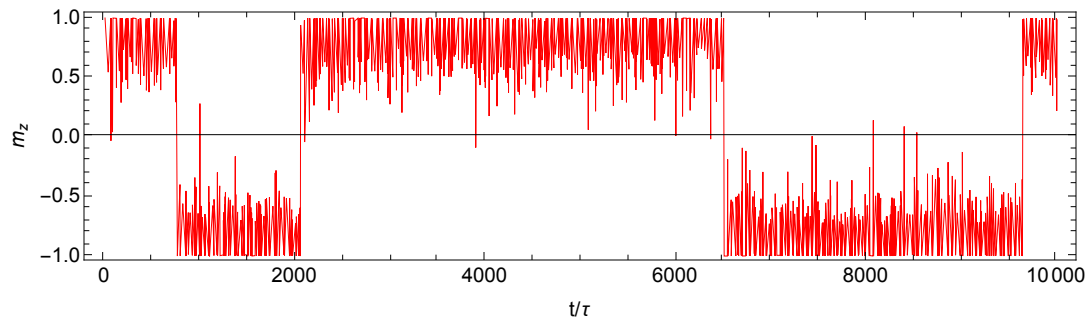| Parameters	Meaning	
θ	$k_B T / M_s^2 V$	Reduced Temperature
λ	$2K / M_s^2$	Reduced Anisotropy Constant
$h_a(t)$	$H_a(t) / M_s$	External field in units of M_s
τ	$1 / \alpha \gamma M_S$	Normalized unit of time
σ	$KV / k_B T = \lambda / 2\theta$	Inverse temperature

For more informartion:
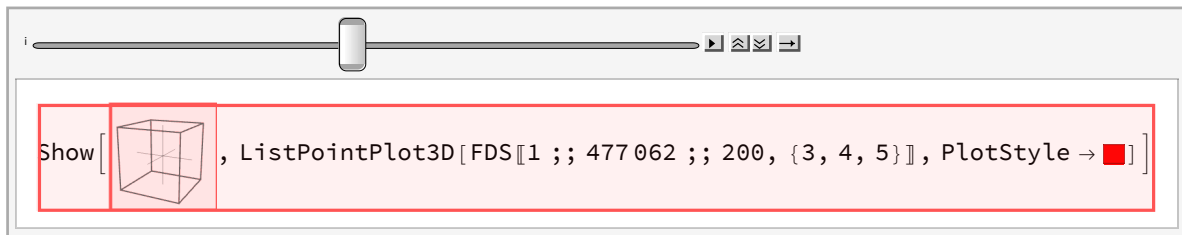
G. T. Landi, *JMMM*, **324** 466 (2012)

G. T. Landi and A. D. Santos, *JAP*, **111** 07D121(2012)
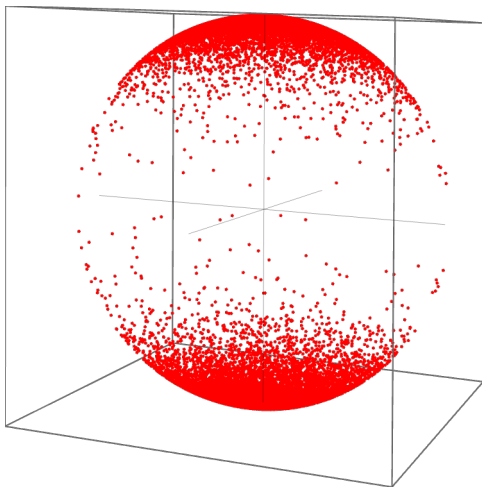
Example: free particle with $\sigma = 15$

◆ m_z vs. time

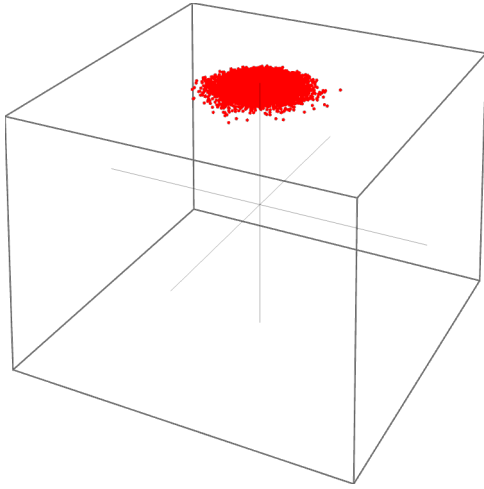◆ 3D view of the magnetization dynamics


```
Show[▨, ListPointPlot3D[FDS〚1 ;; 477 062 ;; 200, {3, 4, 5}〛, PlotStyle → ▪]]
```

◆ Density of points after a long time

What if $\sigma = 100$?

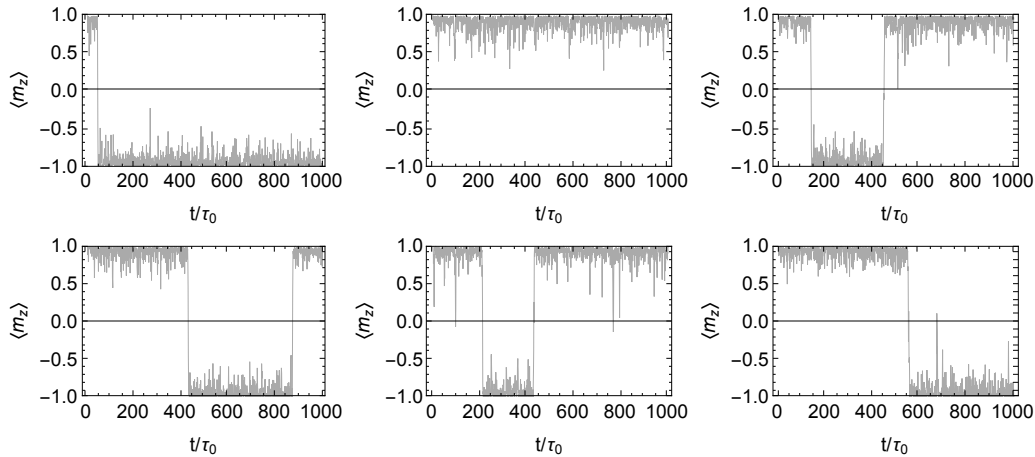*Put the particle in the upper well → It stays there for a long time.*

Averages

◆When we have a sample with many particles, what we observe is the average behavior $\langle m \rangle (t)$.

Example:

◆Suppose we apply a field and align all particles to $m_z = 1$.

◆Then at $t = 0$ we turn the field off and measure what happens.

◆Each stochastic trajectory is very different from the other.

◆We can then take the average.
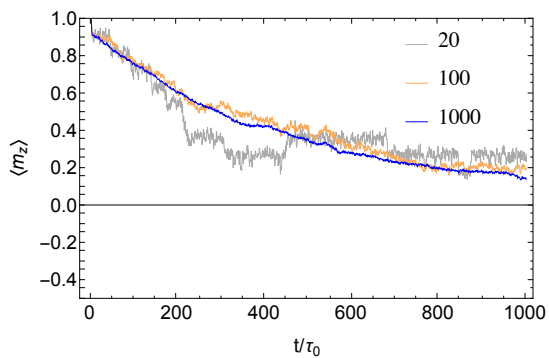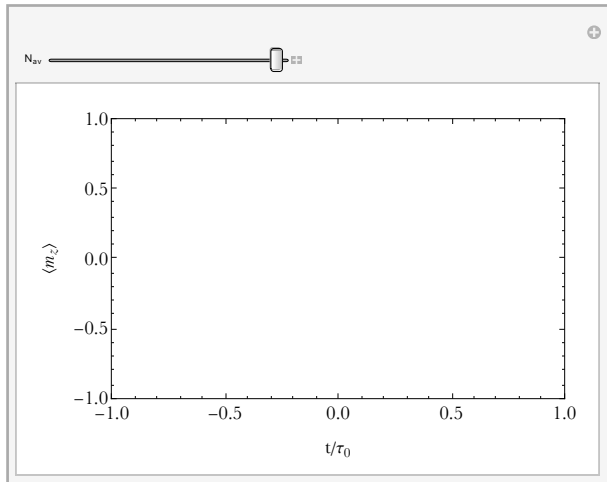
- ◆ The average behaviour is *very* different from the single stochastic realizations.
- ◆ We see that $\langle m_z \rangle$ decays with time toward zero.
- ◆ This means the following:
  - ◆ You start all particles with $m_z = 1$
  - ◆ After a long time they will be randomly distributed between the **north and south poles**.
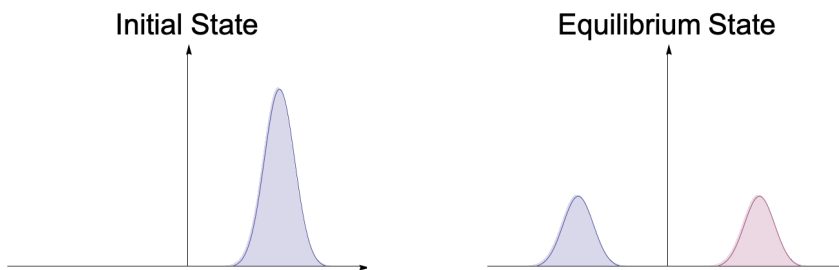
Relaxation time

- ◆ We see clearly from the figure that

$$\langle m_z \rangle \approx e^{-t/\tau}$$

- ◆ Question: *what is the relaxation time τ?*
- ◆ The picture below was taken from a chemistry book.

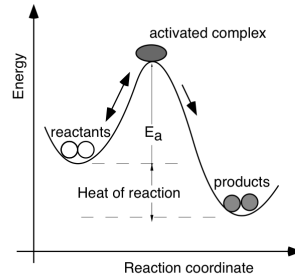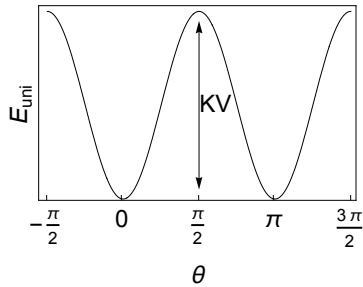Figure 7   Schematic of a reaction via activated complex formation

- This problem is very similar to the *Arrhenius* problem in chemical reactions.
- The result due to Arrhenius is that

$$\tau \propto e^{\Delta E / k_B T}$$

- Thus, in our case we expect

$$\tau \approx \tau_0 \, e^{KV / k_B T} = \tau_0 \, e^{\sigma}$$

- This is called the *Néel relaxation time*.
- The relaxation time grows *exponentially* with σ.

Estimates:

- Usually $\tau_0 \sim 10^{-9}$s. Then

σ	τ (s)
1	10^{-9}
5	10^{-7}
10	10^{-5}
15	10^{-3}
20	10^{-1}
25	10^{2}
30	10^{4}
35	10^{6}
40	10^{8}
45	10^{10}

- At $\sigma = 25$ the relaxation time becomes proportional ~ 100 s, which is the usual time of a magnetometry experiment.
  - Thus, you apply a field to align the particles and before you finish your measurement: POOF, no more magnetization.
- The fact that τ changes exponentially with σ is the reason why in ZFC-FC we say "Hey, its blocked. Hey, its not blocked".
  - This sharp distinction is possible since there is such a rapid change in τ.
- These are the ideas of *superparamagnetism*.

Superparamagnetism is a relative concept.   ← LOOK AT ME!

It depends on the duration of the experiment (or the field sweep).

◆ If your experiment involves a 10 kHz AC field, then the period is $1/10^4 = 10^{-4}$ s.

　◆ For this experiment the particle will be superparamagnetic if $\sigma = 10$ but not if $\sigma = 15$.

◆ In the other extreme, if you make a magnetic tape and want to store your data for 10 y $\sim 10^8$ s you need $\sigma = 40$.

Hysteresis loops

◆ Now let us consider a hysteresis loop using a harmonic field
$$H_a = H_0 \cos (2 \pi f t)$$

◆ The free parameters are the frequency f and the 3 components of $\mathbf{H_0}$

◆ The hysteresis loop of a single particle is very noisy.

◆ But that of many particles is smooth.

Example: $\sigma = 15$, $f / \tau_0 = 10^{-3}$
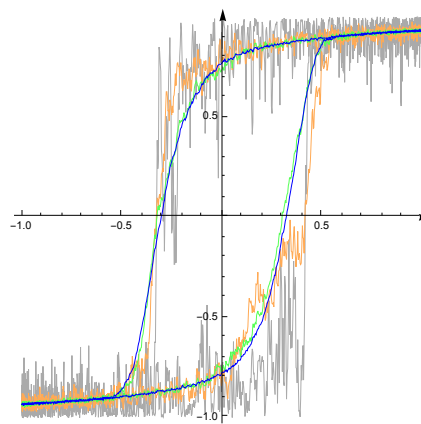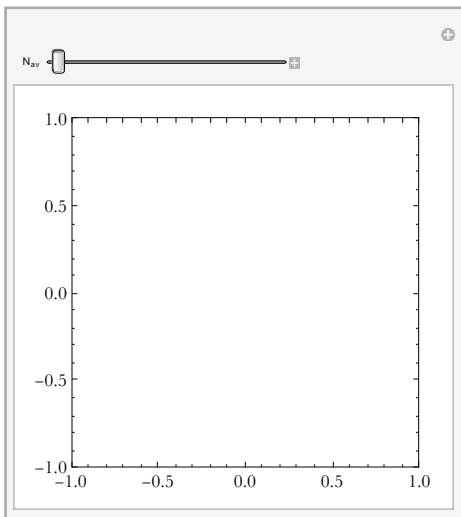
◆ In experiments such as hyperthermia we perform many many many hysteresis cycles in a row.

◆ Hence in this case even the hysteresis loop of a single particle would look smooth, since it would correspond to the average of many cycles.

Example: $\sigma = 3$, $f / \tau_0 = 10^{-3}$
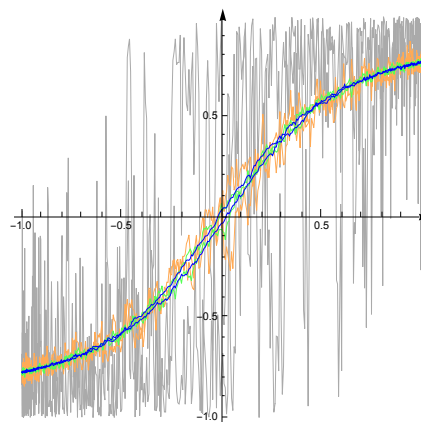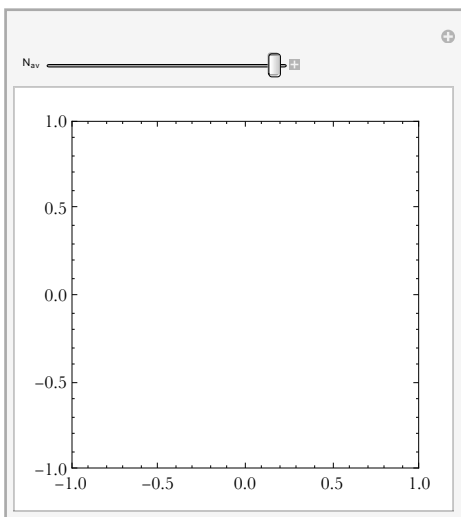
◆ When σ is small (low anisotropy or high temperature) the hysteresis becomes smaller.
   ◆ The behavior tends to that of a regular *paramagnet*.

Equation for the averages

- In almost all practical cases, all we care about is the average $\langle \boldsymbol{m} \rangle (t)$.
- If this is the case, we can try to obtain an equation for the time evolution of each $\langle m_i \rangle (t)$.
- But there is a catch: the equation for $\langle m_i \rangle (t)$ will depend on higher powers, such as $\langle m_i^2 \rangle (t)$.
- Example: when h_a is parallel to z:

$$\tau_0 \frac{d \langle m_z \rangle}{dt} = \left(1 - \frac{1}{\sigma} \right) \langle m_z \rangle - \langle m_z^2 \rangle$$

- Then you say: "OK. Let us find the equation for $\langle m_z^2 \rangle$".
  - Dang! It depends on $\langle m_z^4 \rangle$:

$$\tau_0 \frac{d \langle m_z^2 \rangle}{dt} = \left(2 - \frac{3}{\sigma} \right) \langle m_z^2 \rangle - 2 \langle m_z^4 \rangle + \frac{1}{\sigma}$$

- This *hierarchy* never closes! It goes on and on.

Do not panic.

- This does not mean that this approach is useless.
- What it means is that instead of having one differential equation, we have an infinite number.
- It is a trade

$$\text{1 stochastic DE} \Longleftrightarrow \infty \text{ deterministic DEs}$$

There is no such thing as a free lunch

How to solve using this method:

1. Find a systematic way to write the Eq. for arbitrary $\langle m_i^n \rangle$ for $n = 1,2,3,....$

2. Truncate at a high enough n.

3. Solve the *system* of DEs numerically.

Is it better than the other method?

- Yes! 1000 times better.
- Solving systems of ODEs is very easy!
- We use ~ 1000 equations
  - but 1 curve takes ~ 2 seconds only!
- Now we always get clean curves. No noise!
- This is a *numerically exact method*.
  - We do not depend on statistical averages.

General formula

- Suppose $f(\boldsymbol{m})$ is an arbitrary function of $\boldsymbol{m}$.

◆The general formula is

$$\tau_0 \, \frac{d \langle f \rangle}{dt} = - \left\langle \left[\frac{1}{\alpha} \, \boldsymbol{m} \times \boldsymbol{h} + \boldsymbol{m} \times (\boldsymbol{m} \times \boldsymbol{h}) \right] \cdot \frac{\partial f}{\partial \boldsymbol{m}} \right\rangle + \frac{1}{2 \, \sigma} \left\langle \left(\boldsymbol{m} \times \frac{\partial}{\partial \boldsymbol{m}} \right)^2 f \right\rangle$$

Neat trick: use spherical harmonics

◆Since $|\boldsymbol{m}| = 1$ the dynamics occurs in the unit sphere.

◆Thus we may write $\boldsymbol{m}$ in spherical coordinates with angles θ and ϕ.

◆Then we choose $f(\boldsymbol{m}) = Y_{l,m}(\theta, \phi)$; the spherical harmonics.

◆This gives a hierarchy of equations for $y_{l,m} = \langle Y_{l,m}(\theta, \phi) \rangle$:

$$\tau_0 \, \frac{dy_{l,m}}{dt} = \sum_{l', m'} T_{l,m}^{l', m'} \, y_{l', m'}$$

◆I will not write the full formula. It is very large.

Needless to say: this method **always** *agrees with the stochastic method.*

Example:

◆Dynamic hysteresis for different frequencies f and values of σ.
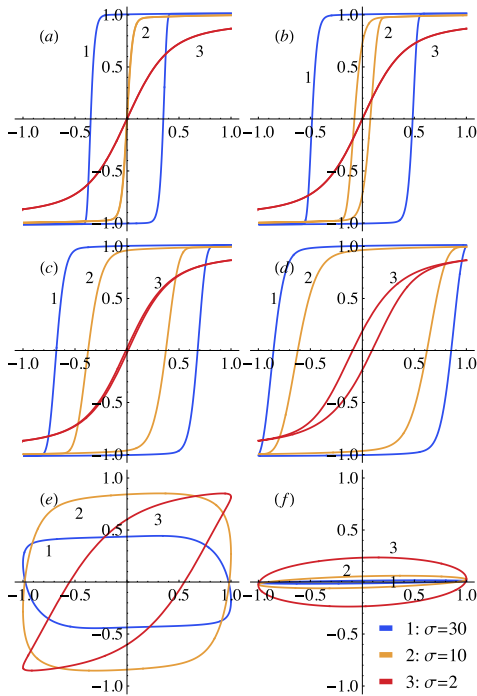
The framework is complete:

◆Now we have a framework where we can simulate *any* experiment involving *single-domain non-interacting particles*.

  ◆Dynamic Hysteresis? Put $H(t) = H_0 \cos(2 \pi f t)$

◆ ZFC-FC: Make the temperature change with time.

◆ FMR: Put $\mathbf{H}(t) = H_z\,\hat{z} + H_x\cos(2\,\pi\,f\,t)\,\hat{x}$

◆ &c. (This is how old books wrote "etc". I think its cool.)

For more information

G. T. Landi and A. D. Santos, *JAP,* **111** 07D121(2012)

G. T. Landi, *JAP,* **111** 043901 (2012)

E. L. Verde, G. T. Landi, ..., A. F. Bakuzis, *JAP,* **111** 123902 (2012)

Hysteresis loops in different conditions

◆ When H_0 is not small and ω is large we get some *weird* loops.

◆ It really depends on the direction between H_0 and the anisotropy axis.

◆ In real samples, each particle has a different easy axis direction so these weird effects "average out".

◆ But some weird stuff remain.

Hysteresis loops for $H_0 \perp$ Ani-Axis

◆ This is the beheavior near resonance. Slight changes in ω completely change the shape of the loop.

◆ Solid curve uses numerically exact method. Points are SLL simulations.

  ◆ As mentioned: they agree perfectly.
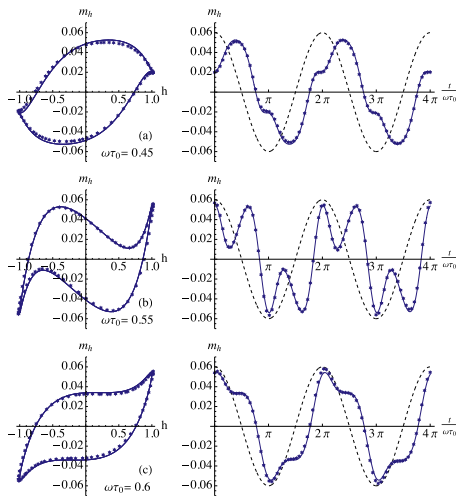
Temperature dependence

◆ Please note: scale is different for each curve.

(a) $\omega\tau_0 = 0.01$ (b) $\omega\tau_0 = 0.1$ (c) $\omega\tau_0 = 0.2$ (d) $\omega\tau_0 = 0.4$
(e) $\omega\tau_0 = 0.55$ (f) $\omega\tau_0 = 0.6$ (g) $\omega\tau_0 = 1$ (h) $\omega\tau_0 = 1.5$

$\sigma = 1$
$\sigma = 2$
$\sigma = 5$
$\sigma = 8$
$\sigma = 10$
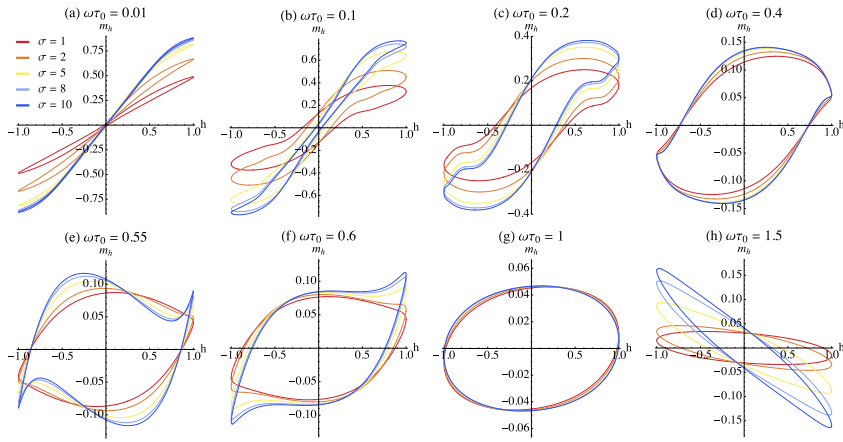
Hysteresis loops for 45°

```
load["Psi45_TrajMini.pdf", 0.4]
```


(a) $\omega\tau_0 = 0.26$

(b) $\omega\tau_0 = 0.28$
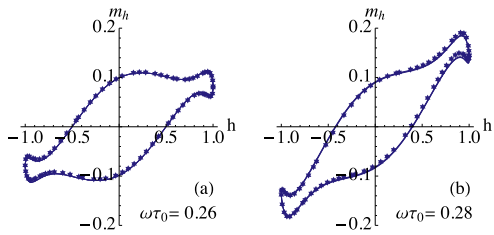
Randomly oriented axis

(a) $\omega\tau_0 = 0.01$

(b) $\omega\tau_0 = 0.1$

(c) $\omega\tau_0 = 0.2$

(d) $\omega\tau_0 = 0.3$

(e) $\omega\tau_0 = 0.55$

(f) $\omega\tau_0 = 1.5$
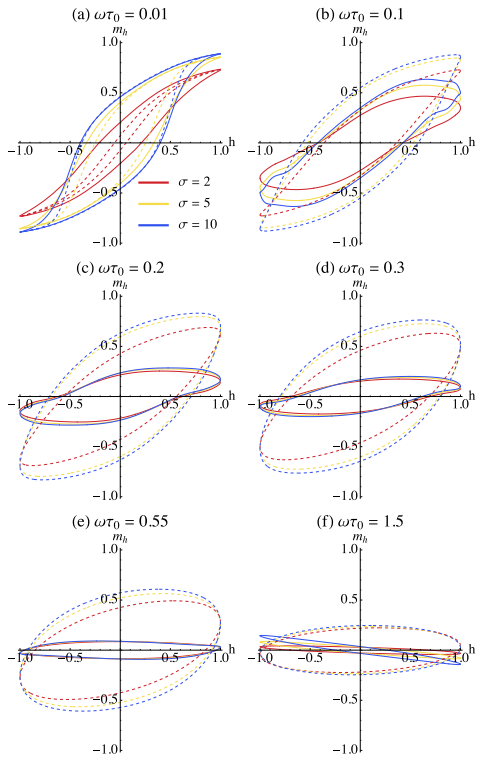
Application: hyperthermia

◆Hyperthermia = dynamic hysteresis + live cell.
◆Hyperthermia = burn the cells with the heat released by particles under AC field.

$H(t) = H_0 \cos(\omega t)$

Cancer cell

Magnetic Nanoparticles

Analogy: microwave

◆A microwave works with a resonance frequency of water.
  ◆But if you use another frequency of an AC *electric* field, you also get dissipation (although alot less).
  ◆This happens because of the water molecule's permanent electric dipole moment that rotate in response to the field.
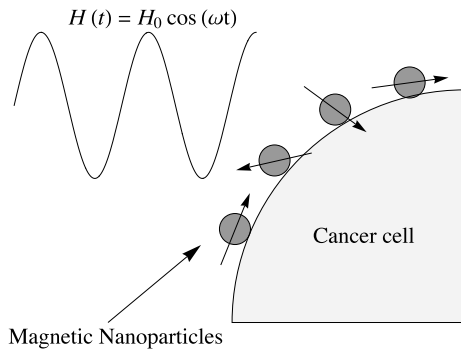 ◆Hence there is friction = heat.
◆Whence: hyperthermia = magnetic microwave (without resonances).

The steps to hyperthermia are:

1. Funcionalize the particle with molecules that adhere to cancer cells.
2. Inject the particles in the blood. They will adhere only at the cancer cells and not at healthy cells.
3. Apply an AC magnetic field $H(t) = H_0 \cos(2\pi f\, t)$.
◆The magnetic moments will respond to the field and this will release heat that will kill the cancer cells.
◆Its a game: you can't use any field you like.
  ◆If H_0 or f are too large eddy currents may form in the body (= bad idea).
◆The challenge is:
  ◆Dissipate the most heat you can.
  ◆Do so with the smallest H_0 and f, and the smallest exposure time possible.

The physics:

◆Heat dissipated depends on the material properties: V, K, M_S, α, etc.
◆How to optimize the materials to improve the efficiency of the treatment.

Collaboration with prof. Andris Bakuzis.

For more information:

E. L. Verde, G. T. Landi, ..., A. F. Bakuzis, *JAP*, **111** 123902 (2012)

E. L. Verde, G. T. Landi, ..., A. F. Bakuzis, *AIP Advances*, **2** 032120 (2012)

G. T. Landi and A. F. Bakuzis, *JAP,* **111** 083915 (2012)

Heat dissipated (review of thermodynamics)

- A hysteresis cycle is a *thermodynamic cycle* (like Carnot's, Otto's, etc.)
- The change of the *internal energy* in an infinitesimal process is

$$dU = dQ + \boldsymbol{H} \cdot d\boldsymbol{\mu}, \quad \boldsymbol{\mu} = \boldsymbol{M}\,V$$

where dQ is the heat released and $\boldsymbol{H} \cdot d\boldsymbol{\mu}$ *is the magnetic work* performed in the system.

- Tip: is it $+\boldsymbol{H}d\boldsymbol{\mu}$ or $-\boldsymbol{H}d\boldsymbol{\mu}$ or $+\boldsymbol{\mu}d\boldsymbol{H}$ or $-\boldsymbol{\mu}d\boldsymbol{H}$?
  - Easy: internal energy is always *Force d(Response)*.
    - ◇ The force is always *intensive* (does not depend on the size of the system).
    - ◇ The response is always *extensive* (increases with system size).
  - + or -? If you increase H, you increase M. So it is +
    - ◇ The only - is for *-pdV*. All other thermodynamic work pairs are +.
- The internal energy U is a function of state:

$$\Delta U = 0 \text{ for a cyclic process}$$

- Hence in a *hysteresis cycle* we get

$$Q = -V \oint \boldsymbol{H} \cdot d\boldsymbol{M} = \text{ area of hysteresis loop}$$

- Big area = big dissipation.
- If you want you can integrate by parts

$$Q = V \oint \boldsymbol{M} \cdot d\boldsymbol{H} = V \int_0^T \boldsymbol{M} \cdot \frac{d\boldsymbol{H}}{dt}\, dt$$

- We usually don't use the heat in one cycle Q,, but the *power* (Joules/second) released in that cycle:

$$S = \text{f}Q$$

- Experimentally it is usual to divide this by the mass m of nanoparticles in order to give an answer in W/g.
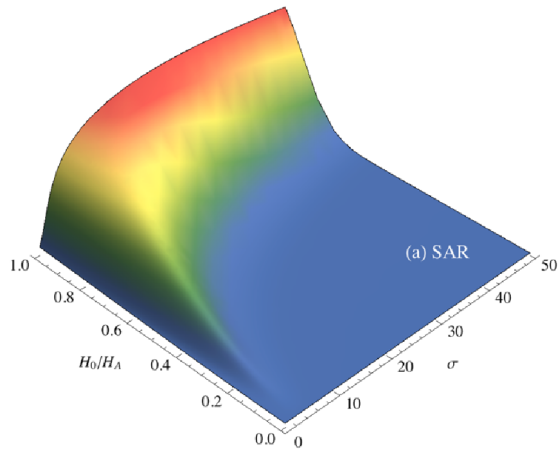 - We call this SAR

$$SAR = S/m = \text{ power dissipated by 1 gram of nanoparticles}$$

For more information

G. T. Landi, *JMMM*, **326** 14 (2013)

Example: SAR vs. H_0 and σ

- The SAR goes up if H_0 goes up.
- The behavior with σ is not so simple.

(a) SAR

Linear response
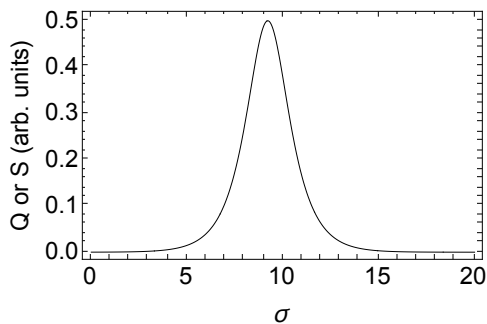
◆ When H_0 is small, the magnetic response is linear in H_0.

 ◆ ∴ $Q \propto H_0^2$ or, what is equivalent, $S \propto H_0^2$

◆ The exact formula for Q is

$$Q = \pi H_0^2 \frac{\omega\tau}{1 + (\omega\tau)^2}, \quad \omega = 2\pi f, \quad \tau = \tau_0 e^\sigma$$

Q and S as a function of σ

◆ As a function of σ both look like this:

◆ There is a value of $\sigma = KV/k_B T$ for which Q (or S) is a maximum.

 ◆ And it is a very sharp maximum!
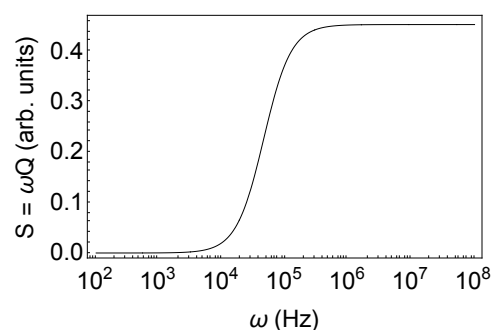
◆ The position of this maximum depends on ω.

$$\sigma_{max} = -\ln(\omega\tau_0)$$

◆ **Challenge**: tune your K and your V to match the max corresponding to the frequency you are using.

 ◆ That will give you a much larger dissipation for a given H_0 and ω.

Q and S as a function of frequency:

◆ If we plot Q or S vs. the frequency ω we get this:
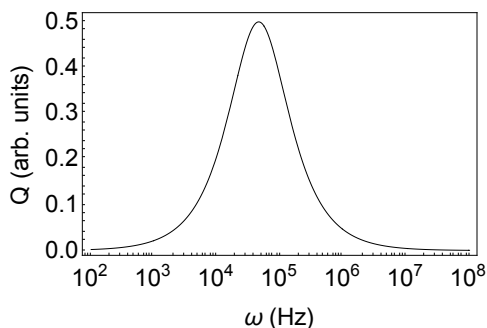
◆ If you increase the SAR you increase increase the dissipation

 ◆ But you also fry your patient!

There is always a size distribution:

◆ There is a distribution of *V and* a distribution of *K*.

◆ But if you assume (and this is reasonable) that both are log-normal, then the distribution of *KV* is also log-normal (this is a property that *only* the log-normal satisfies).

◆ The log-normal distribution is

$$f(x) \; = \; \frac{1}{\sqrt{2\pi}\; \sigma\, x}\; e^{-\log(x/\mu)^2/2\,\sigma^2}$$
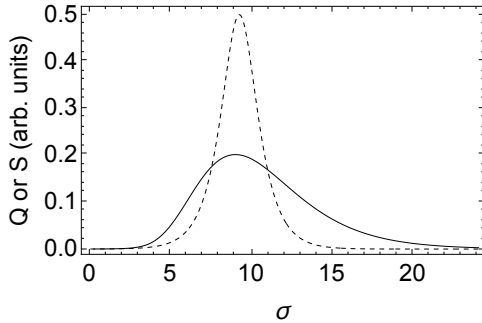
◆ If you are close to the peak, it reduces your dissipation. If you are far away, it increases it.

◆ We have found that using a size distribution gives a good agreement to experiment

```
load["SAR_Experimental.png", 0.4]
```
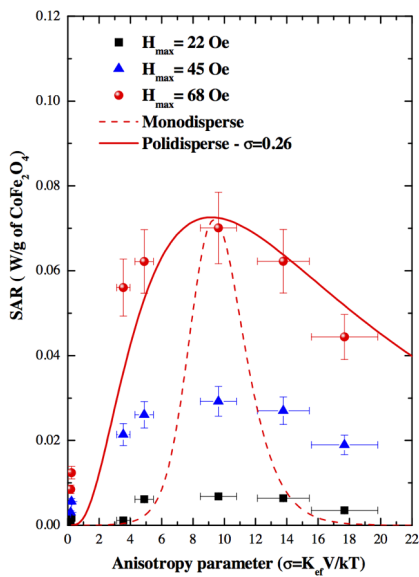
In Q vs. σ we see two regimes
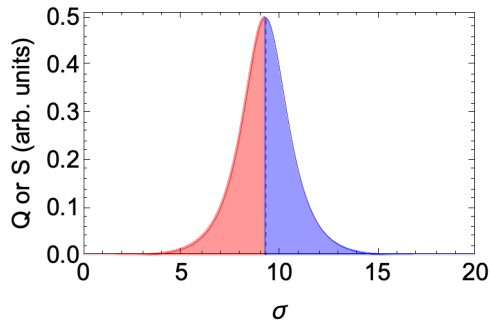
◆ General rule: *alot* of energy is released when the magnetization *flips* from one potential well to the other.

◆ When it is **hot**, the thermal fluctuations make it easy for a tiny field to flip the spins.

  ◆ But the energy released in each flip is small.

◆ When it is **cold**, the spins are "frozen" so a small field doesn't even scratch it.

  ◆ But the energy released in each flip is huge.
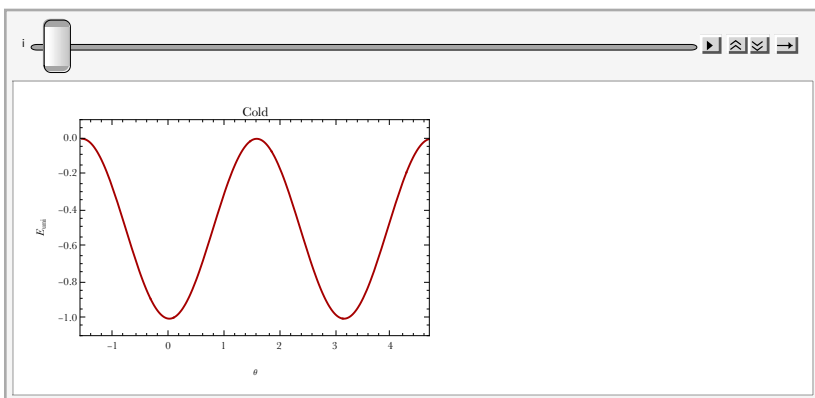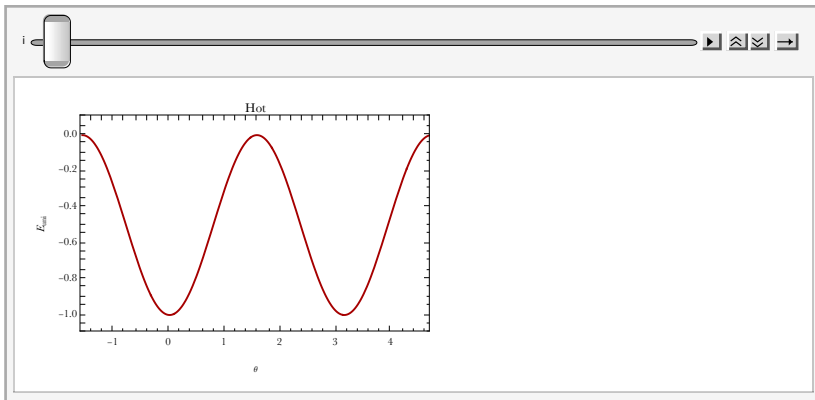
◆ The maximum dissipation represents a *balance* between these two regimes.

The non-linear regime

◆Let us look at experimental data of the SAR vs. H_0

```
img1 = load["SAR_H_1.png", 0.3];
img2 = load["SAR_H_2.png", 0.3];
img4 = load["SAR_H_4.png", 0.24];


txt1 = Text@Style["I. Sharifi, H. Shokrollahi \n and S. Amiri, JMMM 324 (2012)"];
txt2 = Text@Style["D. E. Bordelon, et. al. JAP 109 (2011)"];
txt4 = Text@Style["E. L. Verde, et. al. AIP Advances 2 (2012)"];


Grid[{{img1, img2, img4}, {txt1, txt2, txt4}}, ItemSize → Full]
```
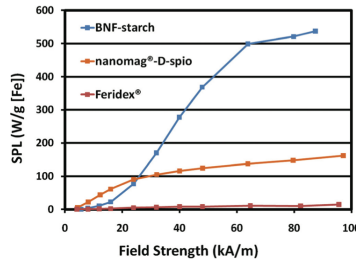

I. Sharifi, H. Shokrollahi
and S. Amiri, JMMM 324 (2012)   D. E. Bordelon, et. al. JAP 109 (2011)   E. L. Verde, et. al. AIP Advances 2 (2012)
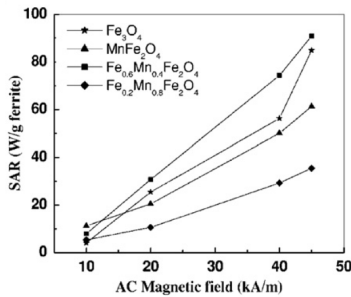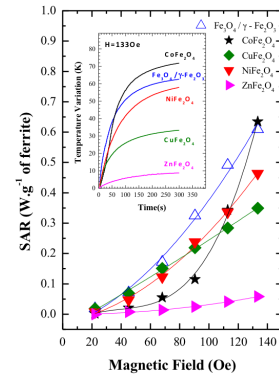
◆What we see is intuitive:

 ◆H_0 goes up → S goes up.
◆But what does all these curves mean?

New concept:

Energy conversion efficiency

G. T. Landi and A. F. Bakuzis, *JAP,* **111** 083915 (2012)

◆When I say "efficiency" you may think about the *"efficiency to kill cancer cells"*.
 ◆If that is the case, the experiment is more efficient when H_0 is larger.
◆But we can also interpret this differently:

> *Hyperthermia is a thermal machine*

 ◆The *energy in* is proportional to H_0^2 (we know this from electromagnetism).
 ◆The *energy out* is Q.
◆We can define the efficiency in converting magnetic energy to thermal energy:

```
Graphics[{

  Arrow[{{0, 0.5}, {1.5, 0.5}}]],
  Arrow[{{3.0, 0.5}, {4.5, 0.5}}]],
  White, EdgeForm[Black], Rectangle[{1.5, 0}, {3.0, 1}]],
  Black,
  Text["Intput Energy ∝ H₀²", {0.7, 0.65}],
  Text["Output Energy = Q", {3.7, 0.65}],
  Text["Magnetic System", {2.25, 0.5}],

  Text["Efficiency := Q/H₀²", {2.25, -0.35}]

}, ImageSize → 650, BaseStyle → 20]
```

Intput Energy $\propto H_0^2$ | Magnetic System | Output Energy = Q
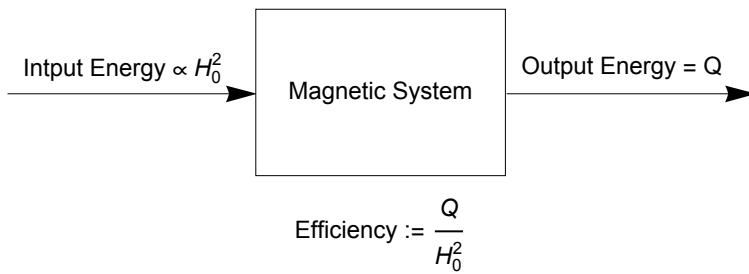
$$\text{Efficiency} := \frac{Q}{H_0^2}$$

◆ In the linear regime Ω is independent of H_0:

$$\Omega = \pi \frac{\omega\tau}{1 + (\omega\tau)^2}$$

Example from "E. L. Verde, et. al. AIP Advances 2 (2012)"

```
load["Efficiency_Experimental.png", 0.4]
```
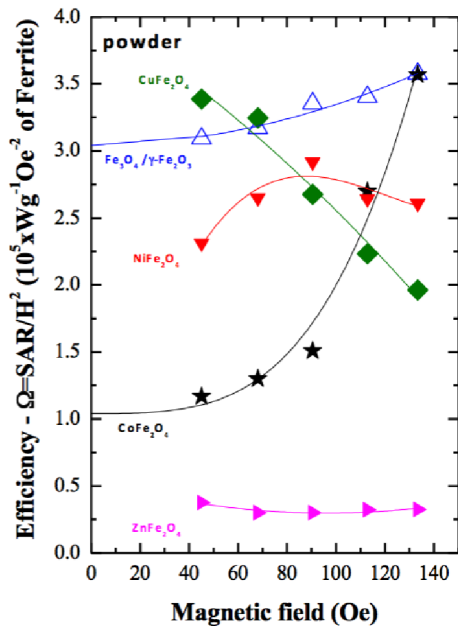
◆ As we increase H_0, the efficiency may either *increase* or *decrease*.

◆ Question: what does this mean?

Heuristic approach: the answer is in the linear response.
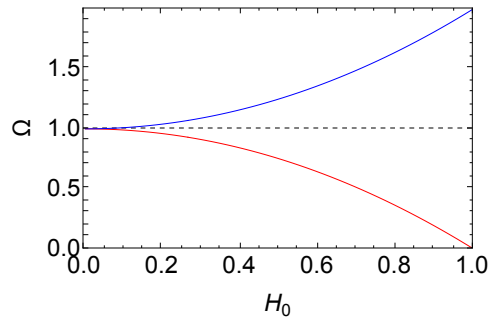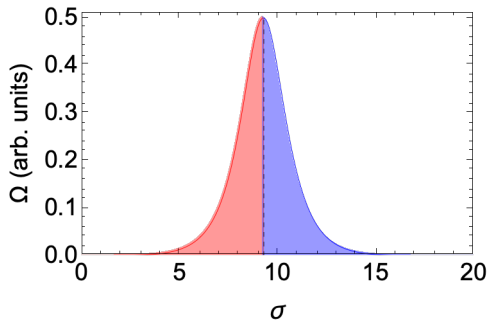
◆ For $\sigma < \sigma_{max}$ it is very easy to flip spins due to the strong thermal aggitation.

   ◆ So increasing H_0 will not make the energy conversion more efficient.

      ◇ Hence the efficiency should decrease.

◆ For $\sigma > \sigma_{max}$ the spins are frozen.

   ◆ So increasing H_0 will unblock the spins and allow them to flip (thus releasing energy)

      ◇ Hence the efficiency should decrease

◆ It makes sense with experiment:
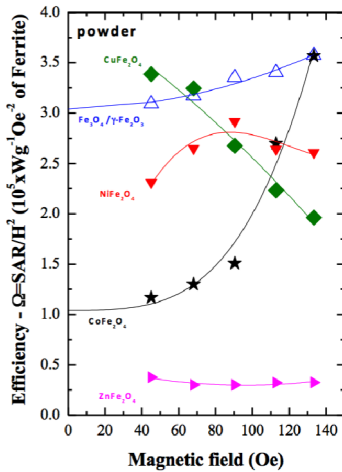
Material	Anisotropy	Efficiency
$CoFe_2O_4$:	Hard	$\Omega \uparrow$
$CuFe_2O_4$:	Soft	$\Omega \downarrow$

Dipolar interaction

◆Everything we have done so far is based on two main assumptions
  ◆The particles are single domain.
  ◆They do not interact with one another.
◆The 1st assumption is reasonable. The 2nd may not be.
◆Each particle produces a magnetic field which acts on other particles.
  ◆This is the *dipolar interaction*.
◆To one degree or another, the dipolar interaction is *always* present.
◆The energy of interaction between two particles, 1 and 2, sepparated by a distance R is

$$U_{\text{dip}} = -\mu_1 \, \mu_2 \left(\frac{\mu_0}{4\,\pi} \right) \frac{\left[3 \, (\boldsymbol{m}_1 . \hat{\boldsymbol{e}}) \, (\boldsymbol{m}_2 . \hat{\boldsymbol{e}}) - \boldsymbol{m}_1 . \boldsymbol{m}_2 \right]}{R^3}$$

where $\boldsymbol{e}$ is the unit vector connecting particles 1 and 2.

*The dipolar interaction is **very** hard to study*

◆It is a long range interaction: *everyone interacts with everyone*.
  ◆This means simulations grow with $O(N)^2$ (computationally expensive).
◆The dipolar interaction has a *reduced symmetry*.

Anti−parallel configuration
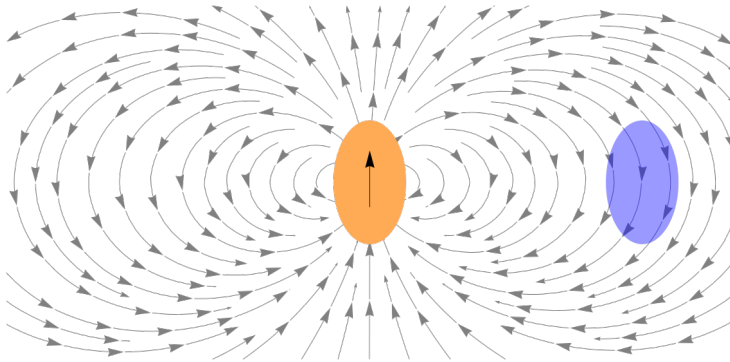
Parallel configuration
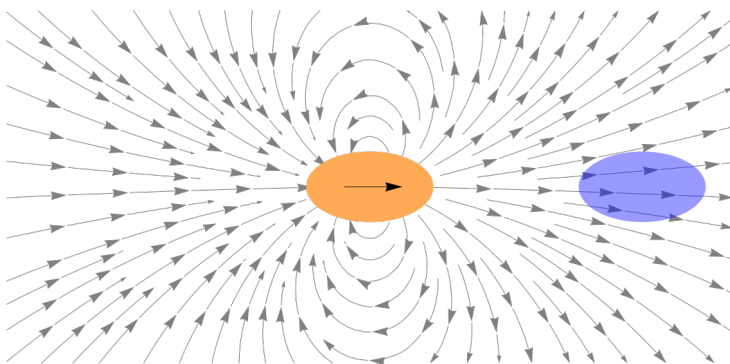
Conclusion

◆ Studying 2, 3, 4, ... particles says *nothing* about the configuration of a ***real system***.

◆ To actually learn about a real system we need to simulate a large number of particles
  ◆ But that is too expensive computationally

◆ You may say "No. That is ≈ what we do in micromagnetism".
  ◆ 1/2 true: in micromagnetism we usually do not include temperature.
  ◆ Here we *must* include temperature.
  ◆ Much more complex.

The mean-field approximation

- ◆I proposed an approximate model to describe the dipolar interaction.
- ◆It is based on the very popular mean-field approximation that is used in ferromagnetism (Curie-Weiss model) and many other areas.

References:

G. T. Landi, *JAP*, **113** 163908 (2013)
G. T. Landi, *Phys Rev. B.,* **89** 014403 (2014)

Review of the Curie-Weiss model

- ◆If we forget about anisotropy for the moment, the energy of a single *isolated* spin is (in reduced units)

$$E = -m\,h$$

- ◆Weiss introduced the idea of a **molecular field** to describe the interaction with the other spins around it.
  - ◆**Idea:** this spins sees, besides the external field h, an average field due to all other spins.
  - ◆This field is proportional to $\langle m \rangle$.

$$E = -m \cdot (h + \lambda \langle m \rangle)$$

- ◆In equilibrium we may compute $\langle m \rangle$; the result is the famous Langevin function

$$\langle m \rangle \;=\; \mathcal{L}\!\left(\frac{h + \lambda \langle m \rangle}{k_B\,T}\right), \quad \mathcal{L}(x) \;=\; \coth(x) - 1/x$$

- ◆This is a self-consistent relation, which we must solve numerically to find $\langle m \rangle$.
- ◆This model predicts a phase-transition from a paramagnetic to a ferromagnetic state.

But this is *not* what we expect for magnetic nanoparticles

- ◆Suppose we start with an infinitely diluted sample and gradually increase the concentration.
- ◆It is known from experiment that the first manifestation of the dipolar interaction is to **increase the anisotropy barrier**.
- ◆Only if the concentration is very very high do we see a correlational order, with spin glass behavior and maybe a ferromagnetic order.
- ◆This is not what the Curie-Weiss model predicts.

The basic idea of a mean-field approximation

- ◆Anyway, let us review what ingredients we need in a mean-field model.
- ◆Instead of focusing on many particles, the idea is to focus on a single (or a small number) of particles.
  - ◆This is good because we can use the techniques already developed to treat a single particle.
- ◆We then want to change the energy of the particle as

$$E \longrightarrow E_{\text{eff}} = E + U_{\text{dip}}$$

where $U_{\text{dip}} \to 0$ in the limit of infinite dilution.

- U_{dip} will depend on the magnetization of the particle: $\boldsymbol{m} = (m_1, m_2, m_3)$.
- But it will also depend on statistical moments of $\boldsymbol{m}$.
  - This is because U_{dip} represents the average interaction with all other particles.
  - These statistical moments have the general form

$$q_{i,j,\dots l} = \langle m_i \, m_j \dots m_l \rangle$$

and U_{dip} may (in principle) be a function of any of the qs.

The result I obtained

- The main result I obtained is this: if you look at one particle, its effective energy will be

$$E_{\text{eff}} = E - k_B T \ln \langle e^{\boldsymbol{m} \cdot \boldsymbol{\xi}} \rangle$$

where $\boldsymbol{\xi}$ is the total dipolar field due to all other particles.

- I treat $\boldsymbol{\xi}$ as a *random variable* so the average $\langle \rangle$ is over all possible values of $\boldsymbol{\xi}$ (which has some underlying distribution that I do not know).
- If we assume there are N particles interacting with our spin then

$$\boldsymbol{\xi} = \sum_{s=1}^{N} \boldsymbol{X}_s$$

$$\boldsymbol{X}_s = \frac{\mu \mu_s}{k_B T} \left(\frac{\mu_0}{4\pi} \right) \left[\frac{3\, \boldsymbol{e}_s (\boldsymbol{e}_s . \boldsymbol{m}_s) - \boldsymbol{m}_s}{R_s^3} \right]$$

Cumulant expansion

- The function $\ln \langle e^{\boldsymbol{m} \cdot \boldsymbol{\xi}} \rangle$ is called the *cumulant generating function of $\boldsymbol{\xi}$*.
- It can be expanded as

$$\ln \langle e^{\boldsymbol{m} \cdot \boldsymbol{\xi}} \rangle = m_i \, \Theta_i + \frac{1}{2!} \, m_i \, m_j \, \Theta_{i,j} + \frac{1}{3!} \, m_i \, m_j \, m_k \, \Theta_{i,j,k} + \dots$$

where the Θ are the cumulants of the distribution

$$\Theta_i = \langle \xi_i \rangle \qquad\qquad \text{(Mean)}$$
$$\Theta_{i,j} = \langle \xi_i \, \xi_j \rangle - \langle \xi_i \rangle \langle \xi_j \rangle \qquad\qquad \text{(Variance)}$$
$$\Theta_{i,j,k} = \langle \xi_i \, \xi_j \, \xi_k \rangle - \langle \xi_i \, \xi_j \rangle \langle \xi_k \rangle [3] + 2 \langle \xi_i \rangle \langle \xi_j \rangle \langle \xi_k \rangle$$

&c.

- The next step is to asssume that each of the $\boldsymbol{X}_s$ above is *statistically independent*.
- We define

$$\eta_a = \left(\frac{\mu \mu_0}{4\pi k_B T} \right)^a \langle \mu^a \rangle \left\langle \frac{1}{R^{3\,a}} \right\rangle$$

- Then we obtain the formulas

$$\Theta_i = 0$$

$$\Theta_{i,j} = \frac{N\eta_2}{5} \, (3 \, \delta_{i,j} - q_{i,j})$$

$$\Theta_{i,j,k} = \frac{2 \, N\eta_3}{35} \, (3 \, \delta_{i,j} \, q_k[3] - q_{i,j,k})$$

&c.

This is confusing, I know.

To make it simpler, let us suppose the external field is $\perp$ to the anisotropy axis

◆ Let us go up to third order and define

$$\frac{N\eta_2}{10} = \nu_2 \, \sigma^2 \qquad\qquad \frac{2 \, N\eta_3}{25} = \nu_3 \, \sigma^3$$

◆ In this particular case we obtain the effective free energy in the form

where the p_i are the averages of Legendre polynomials in m_z:

$$p_1 = \langle P_1(m_z) \rangle = \langle m_z \rangle$$

$$p_2 = \langle P_2(m_z) \rangle = \frac{3 \, \langle m_z^2 \rangle - 1}{2}$$

$$p_3 = \langle P_3(m_z) \rangle = \frac{5 \, \langle m_z^3 \rangle - 3 \, \langle m_z \rangle}{2}$$

&c.

◆ This is what we set out to obtain in the first place:

  ◆ The effective energy now also depends on the averages of m_z.

Results from the mean-field approximation

◆ Our main result is:

$$-\beta E_{\text{eff}} = -\beta E + v_3 \, \sigma^3 (p_1 + p_3 / 14) \, m_z + \left(v_2 \, \sigma^2 \, p_2\right) m_z^2 - \frac{5}{42} \, v_3 \, \sigma^3 \, p_3 \, m_z^3$$

◆ Note that $v_2 \gg v_3$

◆ Hence the first order correction is the red term: **an increase in the anisotropy energy.**

Effect on the relaxation time

◆ Since $p_2 \approx 1$ we have

$$\tau \sim \exp\left(\sigma + v_2 \, \sigma^2\right)$$

◆ This is exactly what I wanted:

　◆ The first order correction of the dipolar interaction is an increase in the anisotropy energy.

◆ This result also relates well to the **Vogel-Fulcher law**:

$$\tau \sim \exp\left(\frac{KV}{k_B(T - T_0)}\right)$$

◆ The term T_0 is a correction due to the dipolar interaction. Expanding

$$\frac{KV}{k_B(T - T_0)} \approx \frac{KV}{k_B \, T} + \frac{KV}{k_B \, T^2} \, T_0$$

◆ T_0 must then be estimated numerically?

◆ Similarly, Shtrikman and Wohlfarth argued that

$$\tau \sim \exp\left[\frac{KV}{k_B \, T} + \left(\frac{\mu H_{\text{dip}}}{k_B \, T}\right)^2\right]$$

where H_{dip} is the average field due to the dipolar interaction

◆ This is precisely the first term in our cumulant expansion.

◆ The difference is that now we know T_0 or H_{dip}!

$$T_0 = \frac{N\mu^2}{10 \, k_B \, KV} \left(\frac{\mu_0}{4\,\pi}\right)^2 \langle \mu^2 \rangle \left\langle \frac{1}{R^6} \right\rangle$$

◆ At least to my knowledge, this is the first time one obtains an explicit formula for T_0 in terms only of system paramaters.

Dynamics and phase transition

◆ We may now put our effective energy in the SLL equation and solve it numerically.

◆ If we include also the third order correction, this model predicts a *phase transition* when the concentration is very high.
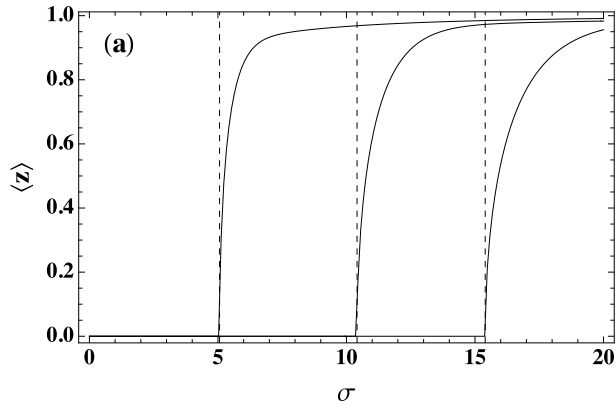
◆ In the critical point the relaxation time diverges.
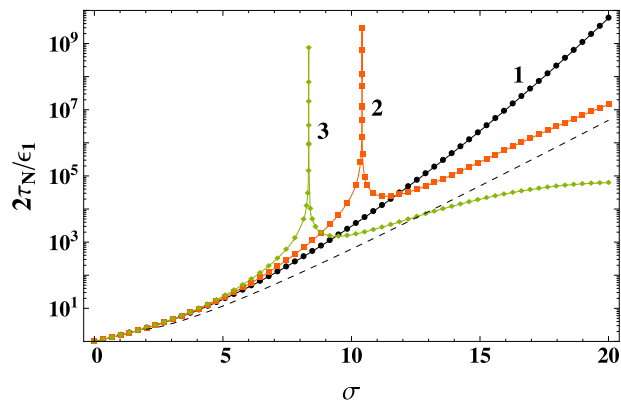
Applications to magnetic hyperthermia

◆Now let us see how the dipolar interaction affects the results of hyperthermia

Linear Regime   Nonlinear Regime

Sorry. γ is the same as ν_2 ● $\sigma = 6$, ■ $\sigma = 8$, ✶ $\sigma = 10$, ○ $\sigma = 12$, ▫ $\sigma = 14$
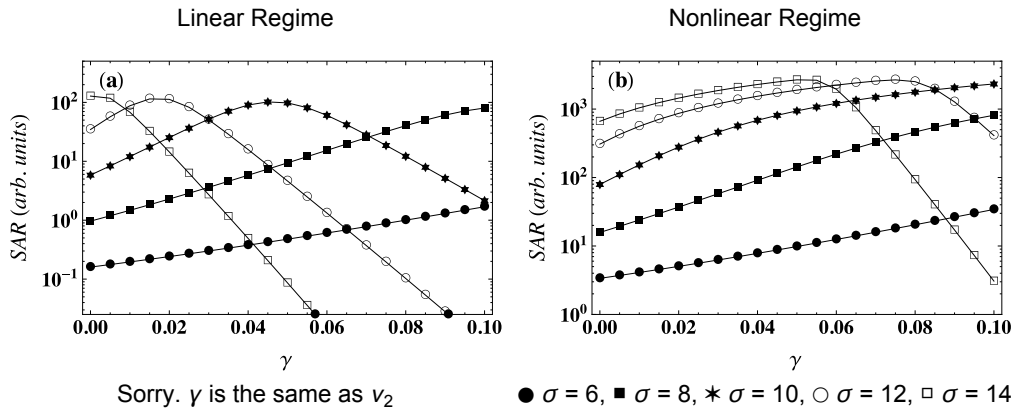
◆The dipolar interaction may either increase or decrease the SAR.

◆It depends on the value of σ of your system.

◆Remember: the dipolar interaction increases the anisotropy barrier.

$$\sigma \;\to\; \sigma + \nu_2\,\sigma^2 \quad \left(\text{or } \sigma + \gamma\sigma^2.\,\text{Sorry again!}\right)$$

◆This increase is huge because it goes with σ^2.

　◆Thus the SAR may be very sensitive to the concentration.

　◆Note the log scale in the graph!

Thank you very much!

Universidade Federal do ABC (Merchandise)

If you want I can tell you more about this new, yet rapidly growing university.

Now graded 5 by CAPES!

Derivation of the effective energy

◆ In this slide I will derive the formula

$$E_{\text{eff}} = E - k_B T \ln \langle e^{m \cdot \xi} \rangle$$

for the effective energy under the dipolar interaction

◆ We start by considering N particles. The total energy can be writen as

$$E_{\text{tot}}(m_1, \ldots, m_N) = \sum_{s=1}^{N} E_s(m_s) + \frac{1}{2} \sum_{s,r} U_{s,r}(m_s, m_r)$$

where E_s is the energy of particle s (Zeeman + Anisotropy) and $U_{s,r}$ is the dipolar interaction between particles s and r:

$$U_{s,r}(m_s, m_r) = -m_s^T \mathcal{D}_{s,r}.m_r$$

◆ Here $\mathcal{D}_{s,r}$ is a 3×3 matrix

$$\mathcal{D}_{s,r} = \mu_s \mu_r \left(\frac{\mu_0}{4\pi} \right) \frac{\left(3 e_{s,r}^T e_{s,r} - I \right)}{R_{s,r}^3}$$

◆ We derive our main result starting from the equilibrium distribution (Gibbs canonical formula)

$$P(m_1, \ldots, m_N) = \frac{e^{-\beta E_{\text{tot}}}}{\int e^{-\beta E_{\text{tot}}} \, dm_1 \ldots dm_N}$$

◆ Let us focus on one particle; say particle # 1.

◆ If we integrate over $m_2, \ldots, m_N$ we get the equilibrium distribution of m_1:

$$P(m_1) = \frac{e^{-\beta E_1(m_1)} \Omega(m_1)}{\int e^{-\beta E_1(m_1)} \Omega(m_1) \, dm_1}$$

where

$$\Omega(m_1) = \int \exp \left\{ -\beta \sum_{s \neq 1} E_s - \frac{\beta}{2} \sum_{s,r} U_{s,r} \right\} dm_2 \ldots dm_N$$

◆ This result is exact: if it were possible to find Ω, it would give the equilibrium distribution of m_1 only.

◆ Now define the dipolar field of particles 2,...N acting on particle 1 as

$$\xi = \beta \sum_{s=2}^{N} \mathcal{D}_{1,s} m_s$$

and change integration variables from $\{m_2, m_3, \ldots, m_N\}$ to $\{\xi, m_3, \ldots, m_N\}$. Then we may write Ω as

$$\Omega(m_1) = \int \Lambda(\xi)$$

Technical functions

Basic function definitions and Style loading

```
SetDirectory[NotebookDirectory[]];
<< "LinLib`";
<< "CustomTicks`";
load[filename_, size_] := Show[Import[filename], ImageSize → Scaled[size]];

SetOptions[Plot, Frame → True, Axes → False,
  BaseStyle → 20, ImageSize → 400, PlotStyle → {Black}];

SetOptions[InputNotebook[],
 DefaultNewCellStyle → "Item",
 ShowCellLabel → "False",
 CellGrouping → Manual,
 FontFamily → "Times",
 DefaultNewCellStyle → {"Text", FontFamily → "Times"},
 BaseStyle → {FontFamily → "Times"},
 MultiLetterItalics → False,
 SingleLetterItalics → Automatic
]
```

SLL Simulation Routine

```
<< "LinLib`";
Clear[SLL];
SLL =
  Compile[{{h0, _Real, 1}, {f, _Real}, {λ, _Real}, {m0, _Real, 1}, {α, _Real}, {θ, _Real},
    {tf, _Real}, {dt, _Real}}, Module[{s, n, m, r, F, G, x, mp, v, FF, GG, ts, hs, R},

    s = N[Sqrt[2 θ α dt / (1 + α²)]];
    n = Floor[tf / dt];
    m = ConstantArray[0., {n, 3}];
    m[[1]] = m0;
    R = RandomVariate[NormalDistribution[0, s], {n, 3}];
    Do[
      (* Partial step *)
      x = m[[i]];
      r = R[[i]];
      v = dt (h0 Cos[2 π f (i - 1) dt] + {0, 0, λ x[[3]]});
            1
      F = - ─ x × v - x × ( x × v);
            α
            1
      G = - ─ x × r - x × (x × r);
            α
      mp = x + F + G;

      (* Full Step *)
      x = mp;
      v = dt (h0 Cos[2 π f i dt] + {0, 0, λ x[[3]]});
             1
      FF = - ─ x × v - x × ( x × v);
             α
             1
      GG = - ─ x × r - x × ( x × r);
             α
      m[[i + 1]] = m[[i]] + 0.5 (F + FF + G + GG) // Normalize;

      , {i, 1, n - 1}];

    ts = dt Range[0, n - 1];
    hs = Norm[h0] Cos[2 π f ts];

    {ts, hs, m[[All, 1]], m[[All, 2]],
                              (h0.#)
       m[[All, 3]], ───────────────────────────────── & /@ m} // Transpose
                    If[Norm[h0] > 10⁻¹², Norm[h0], 1]

  ], CompilationTarget → "C"];
```

```
<< "Simulations.mx";
```

Simulations

Oscillations in the well

```
Clear[h, f, λ, α, θ, tf, dt];
ha = {0., 0., 0.};
f = 0.;
λ = 1.;
α = 1.;
tf = 2 × 10.0³;
dt = 0.05;

θ = 0.09;
OW1 = Table[
    tmp = SLL[ha, f, λ, {0, 0, 1}, α, θ, tf, dt];
    Sign[#〚3〛] ArcCos[#〚5〛] & /@ tmp
    , {10}];

θ = 0.01;
OW2 = Table[
    tmp = SLL[ha, f, λ, {0, 0, 1}, α, θ, tf, dt];
    Sign[#〚3〛] ArcCos[#〚5〛] & /@ tmp
    , {10}];
```

Random Walk

```
RWx = Accumulate@RandomVariate[NormalDistribution[0, 1], 200];
RWy = Accumulate@RandomVariate[NormalDistribution[0, 1], 200];
```

Free Dynamics in the sphere

```
ha = {0., 0., 0.};
f = 0.;
λ = 1.;
α = 1.;
θ = 0.07;
tf = 10 × 10.0³;
dt = 0.01;

FDS = SLL[ha, f, λ, {0, 0, 1}, α, θ, tf, dt];

θ = 0.01;
FDS2 = SLL[ha, f, λ, {0, 0, 1}, α, θ, tf, dt];
```

Relaxation and averages

```
ha = {0., 0., 0.};
f = 0.;
λ = 1.;
α = 1.;
θ = 0.07;
tf = 10.0³;
dt = 0.05;


RA = Table[SLL[ha, f, λ, {0, 0, 1}, α, θ, tf, dt]〚1 ;; -1 ;; 10, 5〛, {1000}];
Do[RAM[i] = Mean /@ Transpose@RA〚1 ;; i〛, {i, 1, 1000}];
```

Hysteresis loop - Cold

```
ha = {Sin[30. °], 0., Cos[30. °]};
f = 0.001;
λ = 1.;
α = 1.;
θ = 0.07;
tf = 10.0³;
dt = 0.05;

HYST = Table[
   tmp = SLL[ha, f, λ, {0, 0, 1}, α, θ, tf, dt];
   H = tmp〚1 ;; -1 ;; 10, 2〛;
   tmp〚1 ;; -1 ;; 10, 6〛
   , {1000}];
Do[HYSTM[i] = Transpose[{H, Mean /@ Transpose@HYST〚1 ;; i〛}], {i, 1, 1000}];
```

Hysteresis loop - Hot

```
ha = {Sin[30. °], 0., Cos[30. °]};
f = 0.001;
λ = 1.;
α = 1.;
θ = 0.3;
tf = 10.0³;
dt = 0.05;

HYST2 = Table[
   tmp = SLL[ha, f, λ, {0, 0, 1}, α, θ, tf, dt];
   H = tmp〚1 ;; -1 ;; 20, 2〛;
   tmp〚1 ;; -1 ;; 20, 6〛
   , {5000}];
Do[HYSTM2[i] = Transpose[{H, Mean /@ Transpose@HYST2〚1 ;; i〛}], {i, 1, 5000}];
```

Saving

```
DumpSave["Simulations.mx",
   {OW1, OW2, RWx, RWy, FDS, FDS2, RA, RAM, HYST, HYSTM, HYST2, HYSTM2}];
```