

Quantitative Magnetization States of Colloidal Systems via Simplified-LLG and Model-based Fitting of Phase Images

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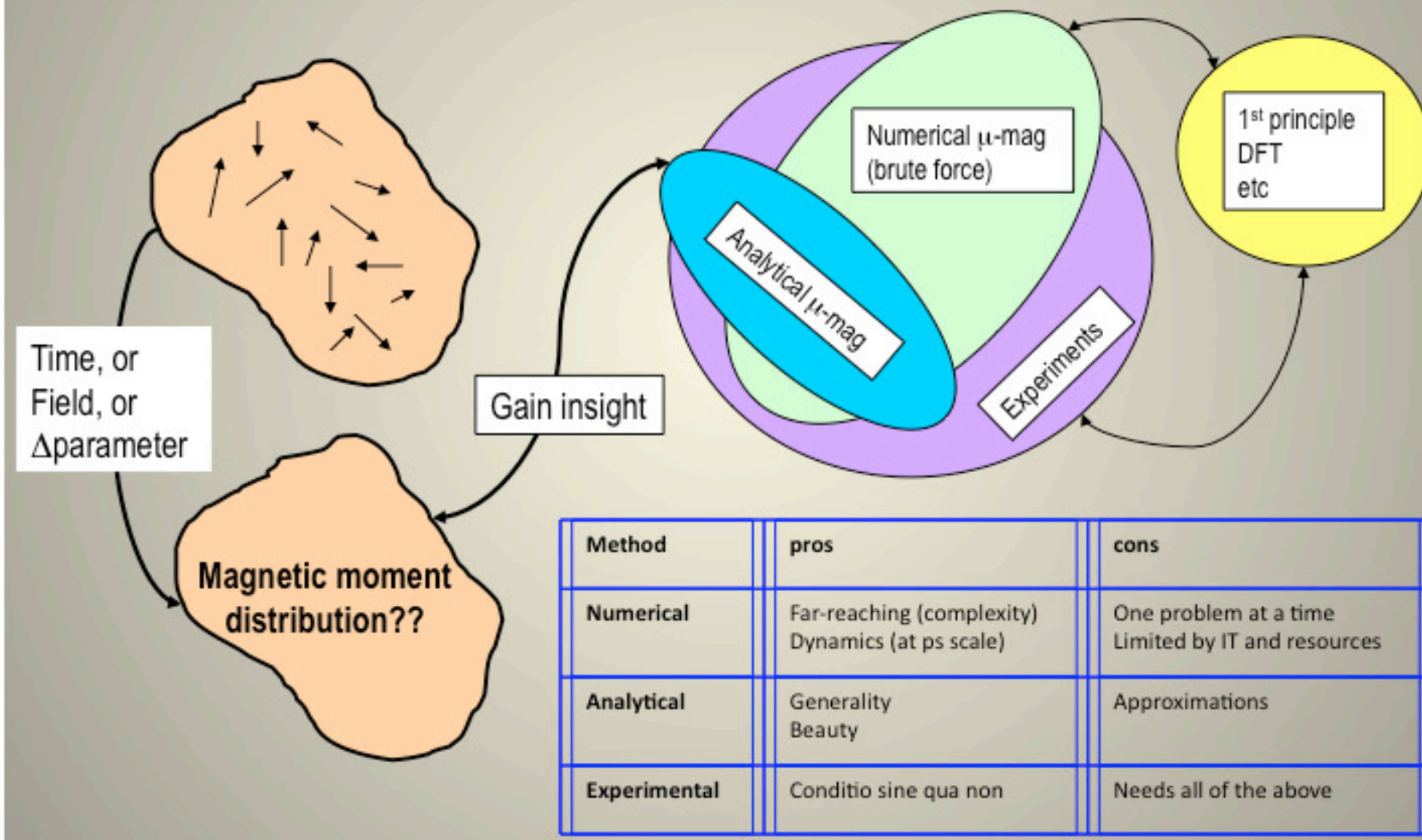
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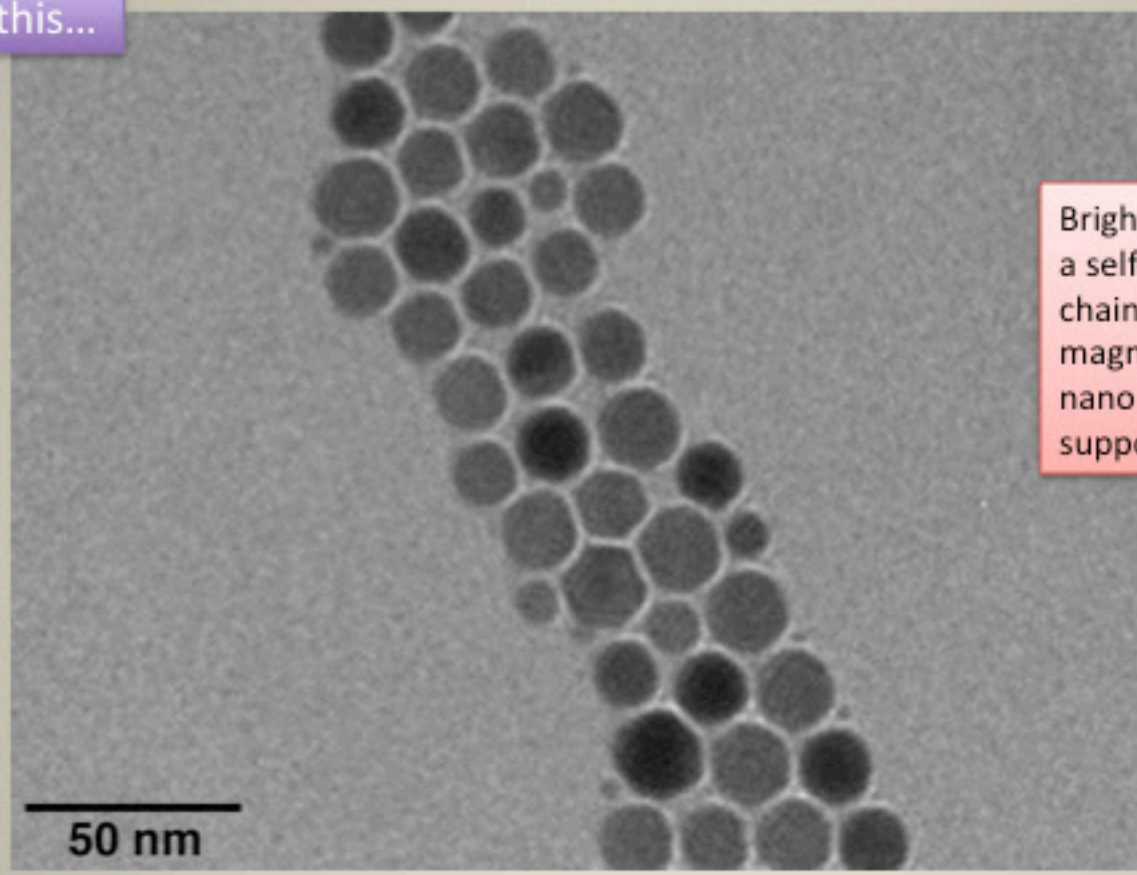
The problem of micromagnetism

What is, and how do we measure, the magnetic state as a function of shape, material and external parameters?



Magnetic colloidal systems

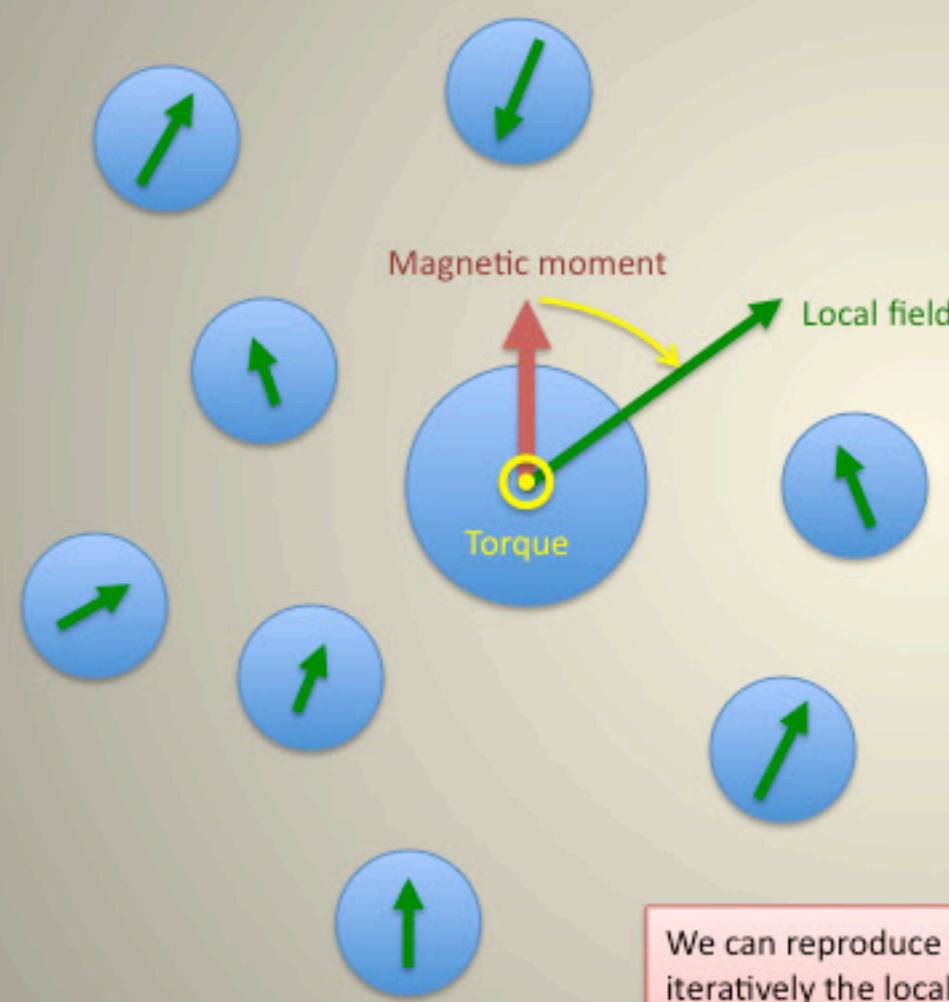
Consider this...



Bright field image of a self-assembled chain of ε-Co magnetic nanoparticles on a C support film.

Each nanoparticle in the chain is a single-domain nanomagnet, free to rotate as it interacts with neighbours and the environment. The key question we want to answer is this: *what is the magnetic moment distribution?* Can we assign an "arrow" to each particle in the chain?

Simplified LLG magnetodynamics



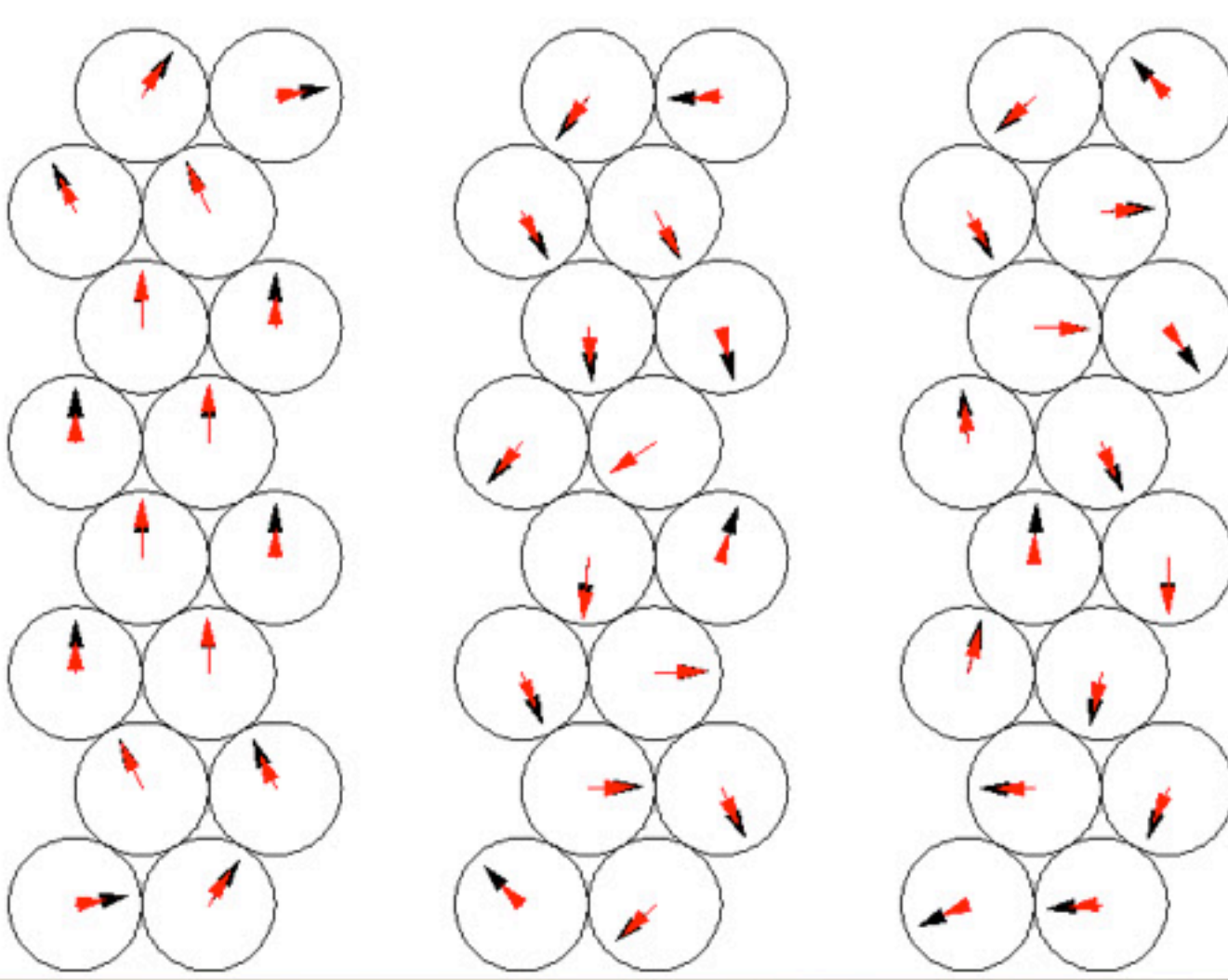
In its simplest form, the LLG equation applied to colloidal systems describes the alignment of moments to the direction of the local magnetic field.

The system relaxes from the initial state (random, or uniform, or assigned arbitrarily) to the final minimum energy state where all torques acting on each and every moment vanish.

We can reproduce the relaxation towards the ground state by calculating iteratively the local field generated by a given moment distribution, from this the torque, from this the new moment distribution, and so forth until the largest residual torque falls below a given threshold

Ground state and local minima

E=-2.45 M=0.86 E=-2.37 M=0.48 E=-2.24 M=0.20



Even for relatively simple colloidal systems, arranged over regular lattices (close-packed in this case), the energy landscape features a vast number of local minima, with energies very close to the ground state.

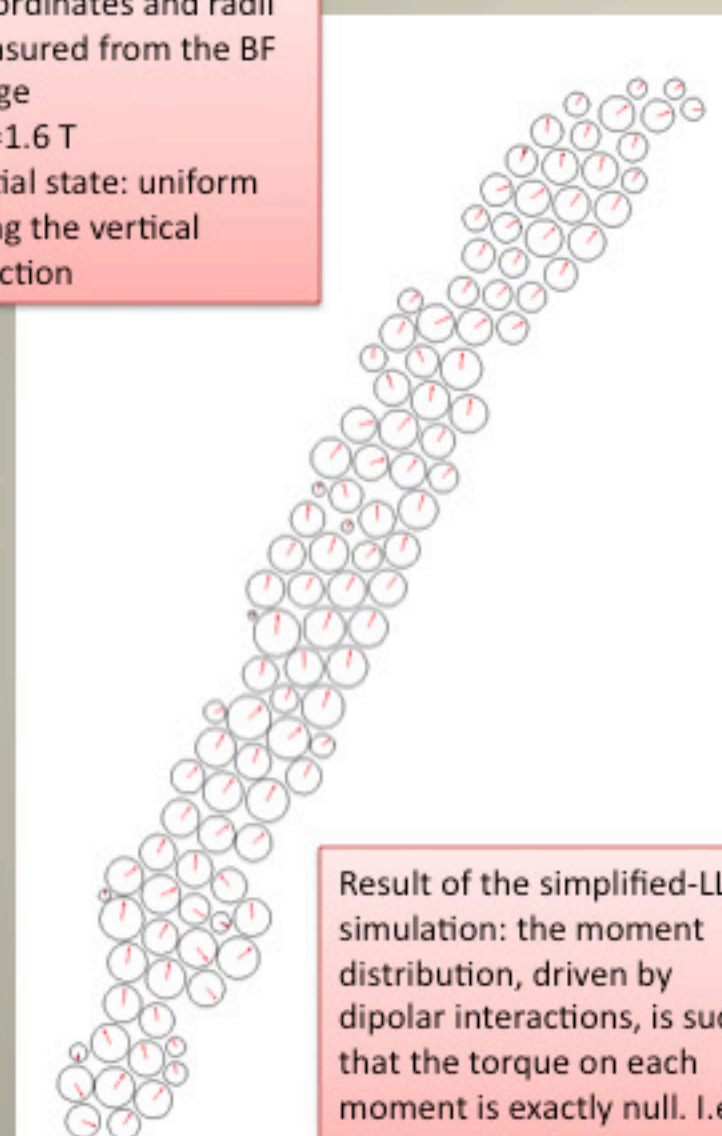
Surprisingly, the torque can vanish within moment distributions that at first sight appear far from equilibrium.

Magnetic history is therefore important in predicting the moment distribution.

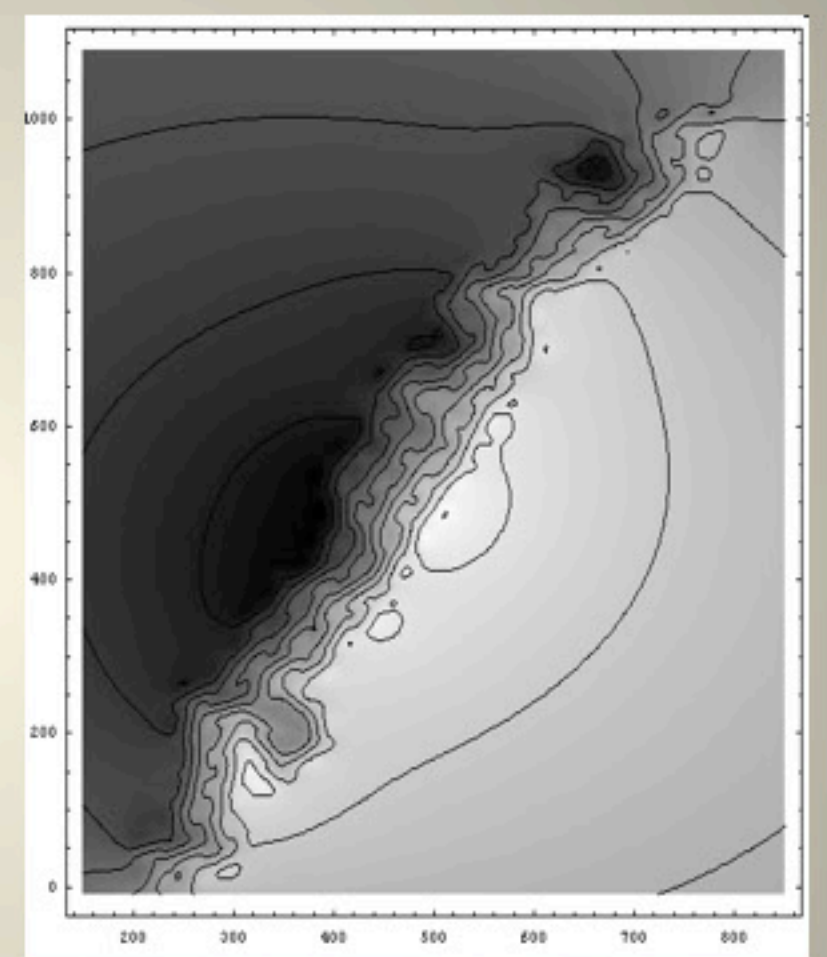
Predicting the moment distribution

Simulation parameters:

- Coordinates and radii measured from the BF image
- $B_0=1.6$ T
- Initial state: uniform along the vertical direction

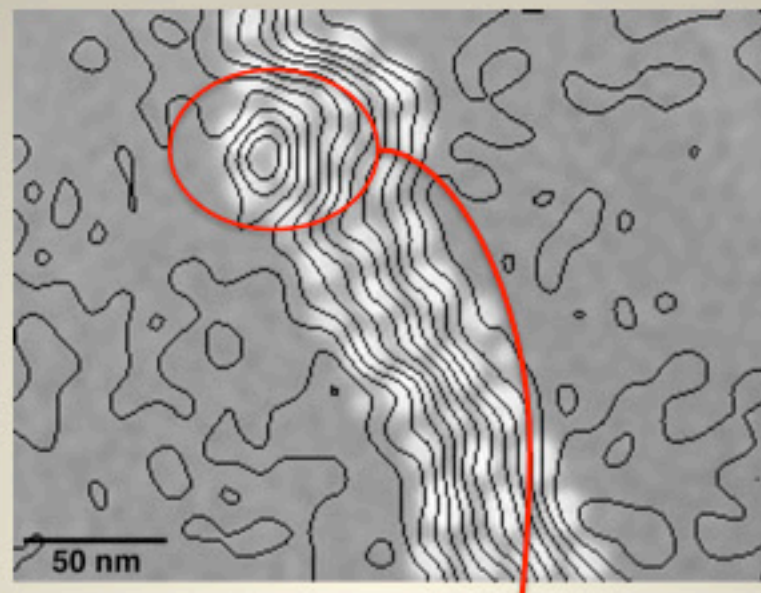
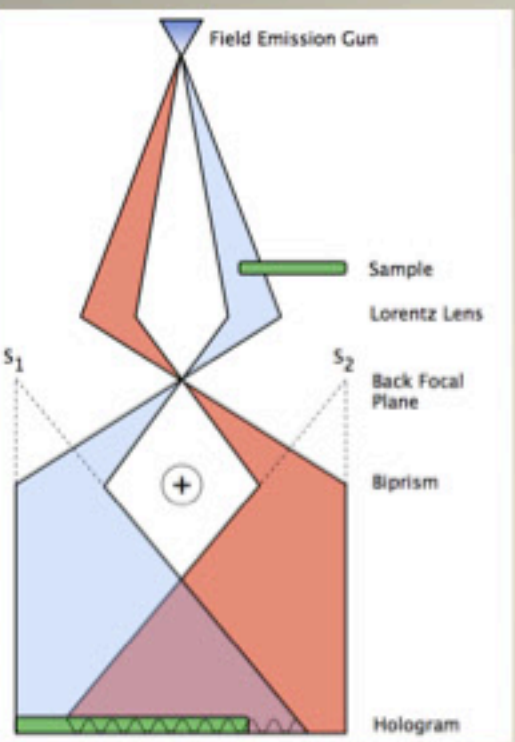


Result of the simplified-LLG simulation: the moment distribution, driven by dipolar interactions, is such that the torque on each moment is exactly null. I.e., static equilibrium.

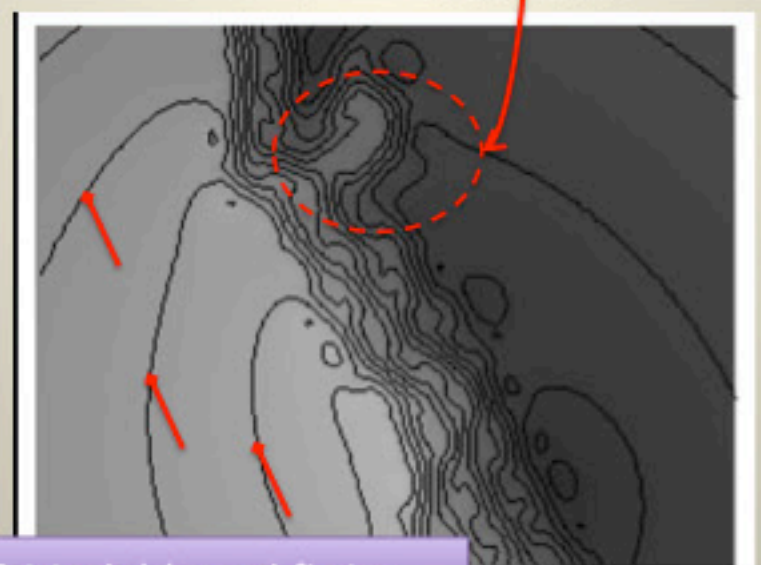


Corresponding simulated phase image, to be compared with the experimental holographic phase shift

Experimental results: Electron Holography



Magnetic contribution to the phase shift, visualized via 0.083 radians phase contours, superimposed to the electrostatic contribution highlighting the nanoparticles positions within the chain.

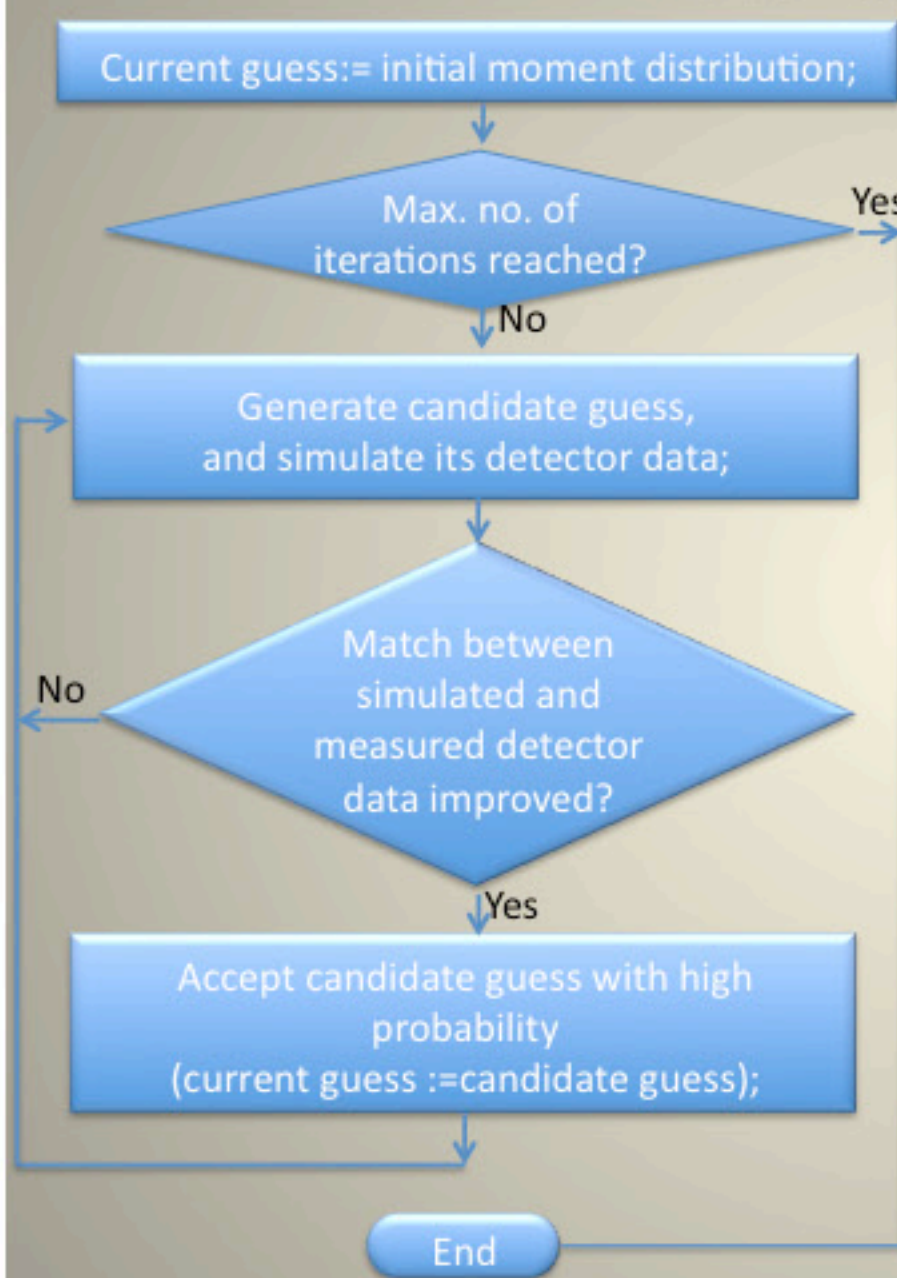


A close-up comparison reveals an satisfactory agreement in terms of number of flux lines and overall appearance. However, fine features are incorrectly captured. Note in particular the vortex-like structure emphasized, which is predicted on the other side of the chain. Furthermore, return flux is visible in the vacuum region (red pointers) surrounding the simulated chain, produced by the poles of the exit poles, with little correspondence to the experimental image.

Electron Holography in the TEM gives us information on the phase modulation of the electron wave, which is proportional to the projected electromagnetic potentials.

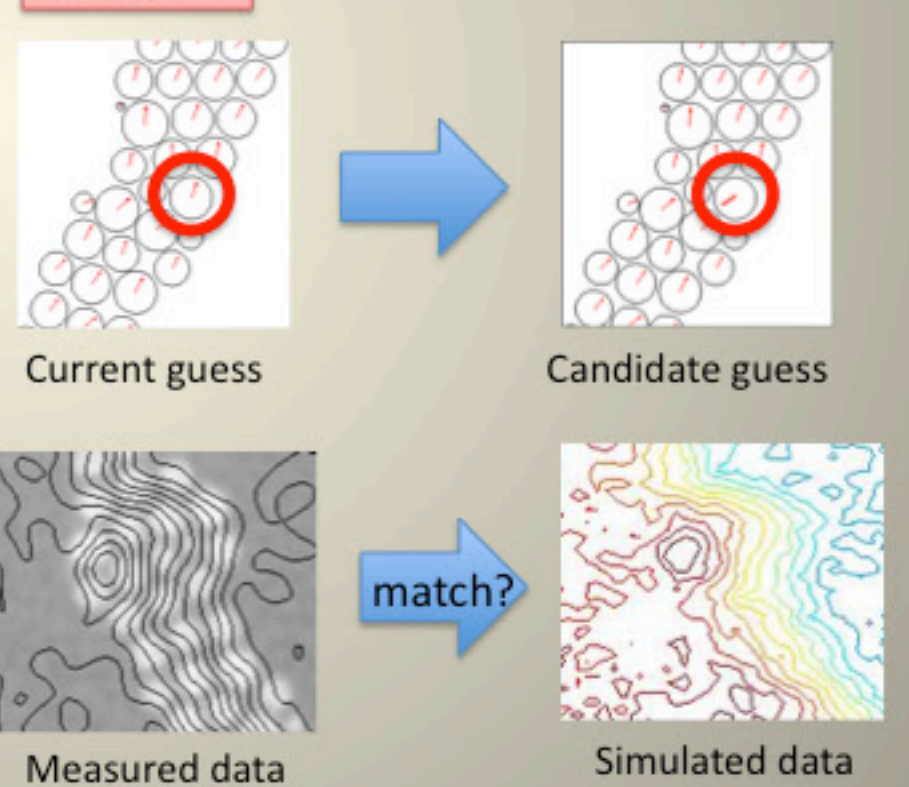
How to make things better? Model-based fitting...

Model-based fitting



The idea is to adopt a Monte-Carlo-type iterative scheme where phase image simulations corresponding to different moment distributions are matched against the data. The discrepancy at any given iteration drives the variation of the moment distribution towards the next attempt, until convergence is achieved.

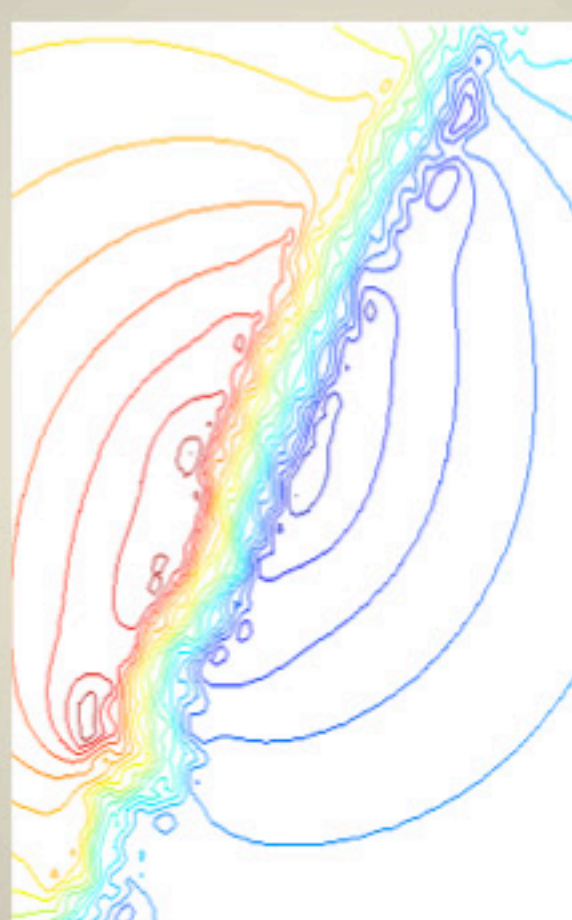
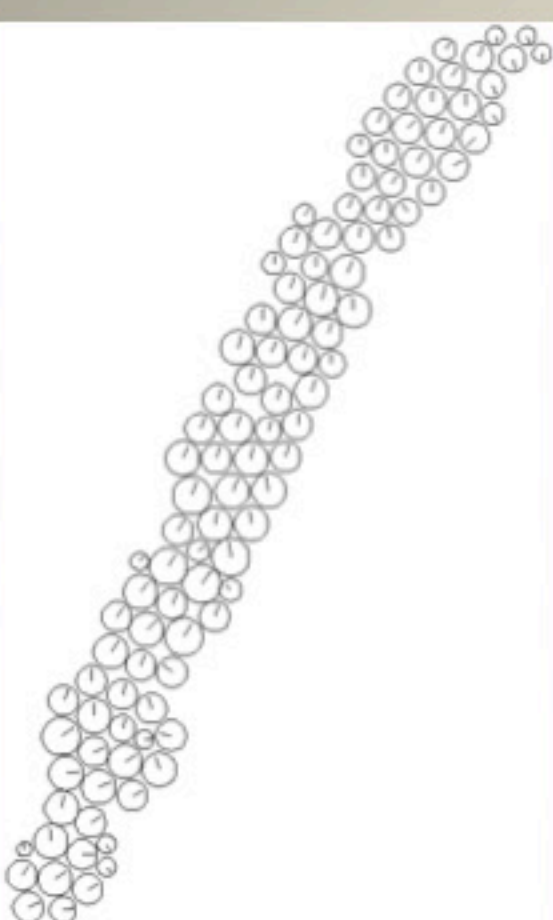
Example:



Measuring the moment distribution

Results

Discussion



The results were obtained on a PC with a 2.66 GHz Intel Core2Duo (1 GB RAM). Computation time: 103s.

The vortex is visible, other features are still wrongly reconstructed.

We observe that the following parameters have a rather **large influence** on the reconstruction results:

- Moments outside the field of view, and
- The estimated radii of the moments.

On the other hand, the initial moment distribution seems to have **no large influence** on the reconstruction.

Reconstructed moments

Corresponding simulated data

Conclusions and perspectives

Conclusions

Measuring the magnitude and orientation of each magnetic moment within an assembly of nanoparticles is a very challenging problem.

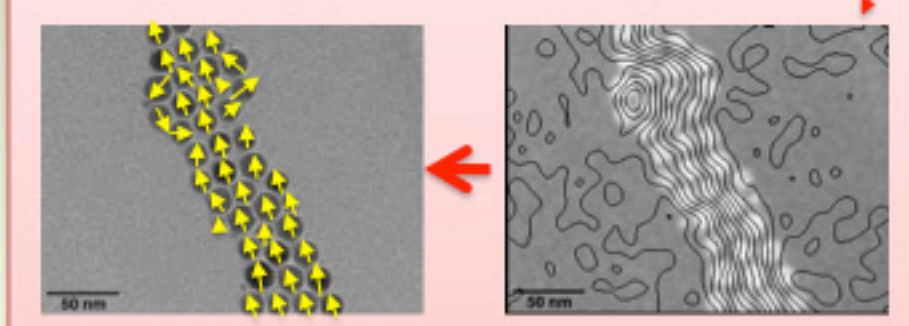
With simplifying assumptions, predicting a likely configuration is simple enough, but simulating via LLG a real system, where NPs may not be spherical, may not lie on the same plane, may interact not only magnetostatically, and may have anisotropies etc., is likely to produce different moment configurations than those observed.

The LLG simulation, however, has value in that it may represent a realistic initial condition that can be fed into more sophisticated algorithms such as model-based fitting.

To increase the level of realism, we may have to carry out 3D simulations where particles are positioned at different heights.

Perspectives

Can we get from projected induction maps like this



to quantitative maps of the moment distribution within colloidal systems, where we can assign an arrow (with correct length) to each and every system component, as sketched on the left?

As a way to achieve this goal, we have started developing a model-based fitting algorithm. While the first results are encouraging, several issues need to be addressed. In particular, we plan to:

- Let the magnitude of each moment be a variable in the Monte-Carlo algorithm
- Use data without moments outside the field of view.

Thank you!