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BIOPHYS

Monte Carlo and Experimental Study of the Magnetic Relaxation of Superparamagnetic Nanoparticle Ensembles

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In this work, the effect of different parameters (size polydispersity of the samples, magnetic anisotropy of the particles, Néel and Brown blocking, and dipolar interaction between the magnetic moments) on the relaxation of superparamagnetic iron oxide nanoparticles (SPIONs) is studied numerically, at thermodynamic equilibrium, using a Metropolis algorithm, and compared with experimental data obtained on a Vibrating Sample Magnetometer.

I. Context

- The base theory for modeling the magnetic relaxation of SPIONs is that of Paul Langevin[1].
- It describes an ideal case and does not consider
  - The polydispersity of sizes in a real sample;
  - The particles' magnetic anisotropy;
  - The Néel and Brown blocking in the relaxation process at low temperatures[2];
  - The dipolar interaction between magnetic moments (which in biological media, where the particles tend to form clusters[3], might not be negligible).
- Those effects can be studied analytically only separately. Therefore, to integrate them in a coherent experiment-like model, Monte Carlo simulations are a powerful tool.

II. Simulations

- Based on a Metropolis algorithm, whose energy term comprises
  - the Zeeman interaction with the external field

$$E_i = -\vec{\mu_i} \cdot \vec{B_{ext}} \ ;$$

  - the magnetic anisotropy of the particle

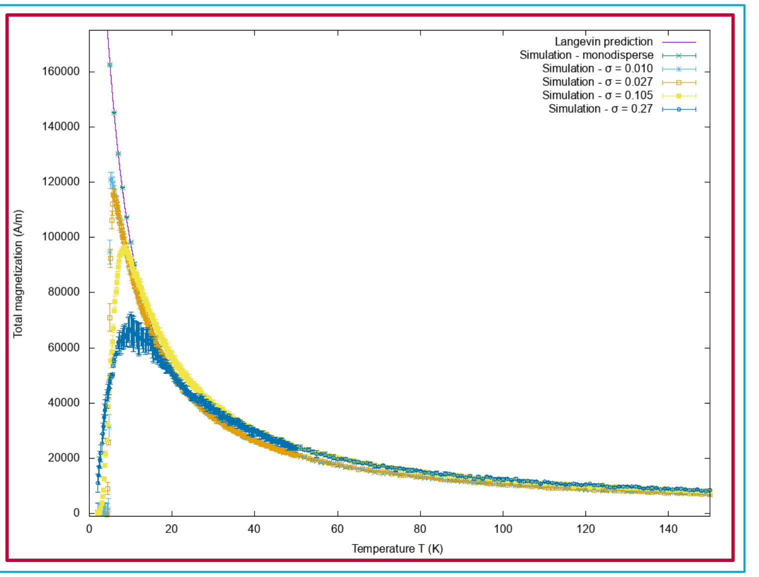
$$E_i = K_i V_i \sin^2 \theta_{i/ext} \ ;$$

  - the dipolar interaction with its closest neighbors

$$E_i = -\frac{\mu_0}{4\pi} \sum_{j,CN} \left[\frac{3\,(\vec{\mu_i}\cdot\vec{r_{ij}})(\vec{\mu_j}\cdot\vec{r_{ij}})}{r_{ij}^5} - \frac{(\vec{\mu_i}\cdot\vec{\mu_j})}{r_{ij}^3} \right].$$

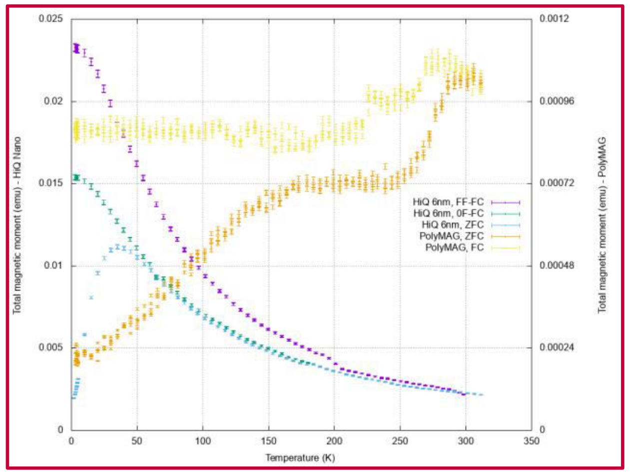
- Adapted to account for Néel and Brown blocking[2,4], since those time-dependent effects are not explained by free energy minimization.
- Particle radii (and therefore, moments) are distributed (following either a lognormal or a normal size distribution).

Simulated ZFC curves of 1000 particles with <r> = 3nm. The radii of the particles in the simulation exhibit a lognormal distribution, whose parameters are varied. B_{ext} = 5mT, K = 13600 J/m³, the anisotropy axises are randomly distributed.



III. Experiments

- Data obtained on a Cryogenics Vibrating Sample Magnetometer.
- 3 samples:
  - 6-nm HiQ Nano particles in chloroform (isolated particles);
  - 12-nm HiQ Nano particles in chloroform (isolated particles);
  - PolyMAG particles (developed for magnetofection) in water (heavily clustered).
- Different types of curves studied:
  - M-H curves of the magnetization as a function of the external field.
  - Zero Field Cooling (ZFC) and Field Cooling (FC) curves;
  - FC curves started below the sample freezing point; i.e the sample is frozen under zero field before applying the 5mT field, cooling it down to 2K and recording the magnetization .
- The notable difference in the FC curves obtained from the two different protocols suggests an effect on the FC curves of the Brown relaxation, which should be studied numerically.
- The PolyMAG particles exhibit a very peculiar ZFC curve, which remains to be explained. Numerical simulation could be the right tool for that goal.



Magnetic measurements at 5mT from a two samples, obtained following various experimental protocols: ZFC, classic FC (FF-FC), and FC after freezing the sample under zero field and then switching on the field (0F-FC).

IV. Conclusion and prospects

- The experiments suggest an important contribution of the Brown relaxation mechanism.
- Simulations are currently being conducted to investigate the importance of the Brown relaxation on the shape of the ZFC and FC curves.
- In parallel, the impact of the various physical parameters (size distribution, dipolar interaction, magnetic anisotropy, …) on the M-H, FC and ZFC curves is being systematically studied.
- The next step will be to investigate the magnetic behavior of particle clusters.
- The final goal of the simulation is to be able to replicate and quantitatively explain the experimental results.

[1] P. Langevin, *J. Phys. Theor. Appl.*, 1905, *4(1)*, 678
[2] W.F. Brown, Jr, *Physical Review*, 1963, *130(5)*, 1677
[3] M. Lévy, C. Wilhelm, et al., *Nanoscale*, 2011, *3*, 4402
4 F. Tournus, A. Tamion, *JMMM*, 2011, *323*, 1118

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