# Monte Carlo and Experimental Study of the Magnetic Behaviour of Superparamagnetic Nanoparticles

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The field-dependant magnetization of superparamagnetic iron oxide nanoparticles is probed through Monte Carlo simulations and compared to experimental results obtained on a Cryogenics Vibrating Sample Magnetometer. The particles' size distribution is obtained by fitting the curves and compared to the actual simulated or TEM-evaluated experimental distribution.

## I. Context

- The naive description of the magnetic behaviour of superparamagnetic iron oxide nanoparticles, usually used to fit magnetization curves, is that proposed by Paul Langevin in 1905.[1]
- For a sample with a size dispersion $f(R)$ :

$$M(B) = \frac{1}{V_{tot}} \int f(R)\, \mu(R) L\left(\frac{\mu(R)B}{kT}\right) dR$$

- However, that model does not take into account the particles' magnetic anisotropy, the magnetic dipole-dipole interactions, or the underlying relaxation mechanisms, which are all complex to take into account theoretically.

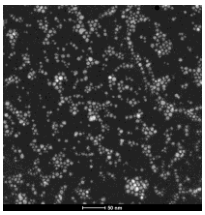
## II. Methodology of the simulations

- A Metropolis algorithm based on the Hamiltonian :

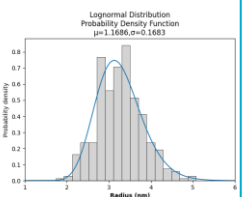
$$E = -\vec{\mu} \cdot \vec{B} - KV\overrightarrow{1_A} \cdot \overrightarrow{1_B} - \frac{\mu_0}{4\pi}\left[\frac{3(\overrightarrow{\mu_i} \cdot \overrightarrow{r_{ij}})(\overrightarrow{\mu_j} \cdot \overrightarrow{r_{ij}})}{r_{ij}^3} - \frac{\overrightarrow{\mu_i} \cdot \overrightarrow{\mu_j}}{r_{ij}^5}\right]$$

- Two relaxation processes are possible :
  - Néel relaxation (i.e. internal reorientation of the magnetic moment);
  - Brown relaxation (i.e. rotation, modeled by a reorientation of the particle's anisotropy axis).
- Dipole-dipole interactions are considered only for particles within a certain cutoff radius of one another.

## III. The experimental sample

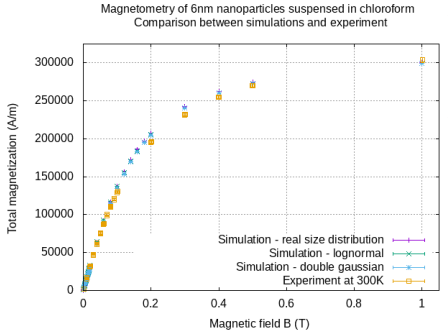


19,7 mgFe/mL (chloroform)




- The radius of 345 particles was evaluated from the TEM imaging of the sample.
- The size distribution was then input in the simulation algorithm either
  - Directly (i.e. the actual particle sizes were input);
  - Through a lognormal fitting of the histogram;
  - Through a gaussian peak fitting of the histogram (satisfactory with two peaks).

## IV. Results




- The simulations compare quantitatively well with the experiments.
- For such small particles ($R_0$ around 3,21 nm), our simulations show no impact of magnetic dipole-dipole interactions at any temperature and any concentration (up to 470 mg /mL with particles placed on a grid for maximum compacity).
- The Brown relaxation process does not contribute significantly to the magnetization curve at 300K either, nor anisotropy (as long as the easy axes of the particles in the sample are randomly oriented).
- Those conclusions are however not valid anymore when even a few bigger particles are present in the sample; the Langevin model should only be used to fit the magnetization curves of small particles exhibiting a narrow size dispersion.

## V. Conclusion and limitations of the methodology

- Our simulation model reproduces well the experimental magnetization curve of a liquid sample of small particles exhibiting a narrow size distribution, and no clustering at 300K.
- Magnetic dipole-dipole interactions do not impact the magnetization curve. This is consistent with studies led on particle clusters. [2]
- It should be noted that the Metropolis algorithm is built to study the equilibrium properties of systems [2], and can not take into account as is the dynamical effects in the particles' magnetization, such as the Néel and Brown blocking of samples at low temperatures.

[1] P. Langevin, Sur la théorie du magnétisme, Journal de physique théorique et appliquée 4 (1) (1905) 678–693. doi:10.1051/jphystap:019050040067800.
[2] J. Londoño-Navarro, J. C. Riaño-Rojas, E. Restrepo-Parra, Competition between anisotropy and dipolar interaction in multicore nanoparticles: Monte carlo simulation, DYNA 82 (194) (2015) 66–71. doi:10.15446/dyna.v82n194.44297.
[3] N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, E. Teller, Equation of state calculations by fast computing machines, The Journal of Chemical Physics 21 (1953) 1087–1092. doi:10.1063/1.1699114.

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