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Investigation of the MPI Signal's Dependency on Ferrofluid Concentration

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Abstract

LANGEVIN's single particle model (**SPM**) yields a *linear* relation between magnetic particle concentration c and higher harmonic amplitudes in magnetic particle imaging (**MPI**). In reality, the MPI signal is *nonlinearly* related to c, as demonstrated via simulation and experiment. Depending on the actual field of application, this may have severe impact on MPI.

Inadequacy of paramagnetic theory

The SPM is a well-established ferrofluid magnetization theory in the field of MPI [1]. Due to its paramagnetic nature, it describes *noninteracting* magnetic particles. Therefore, it is valid only in the limit of particle concentration $c \to 0$. In a real ferrofluid, particles interact magnetically. The smaller the distance between single magnetic moments, the more they support each other in the process of alignment to an external magnetic field H. Hence, the magnetization curve $M(H)$ *nonlinearly* depends on c: increasing c leads to a steeper curve; saturation is reached for weaker H. This effect is ignored by the SPM. The impact of particle interaction can lead to drastic deviations from the SPM, as visualized in Figure 1.

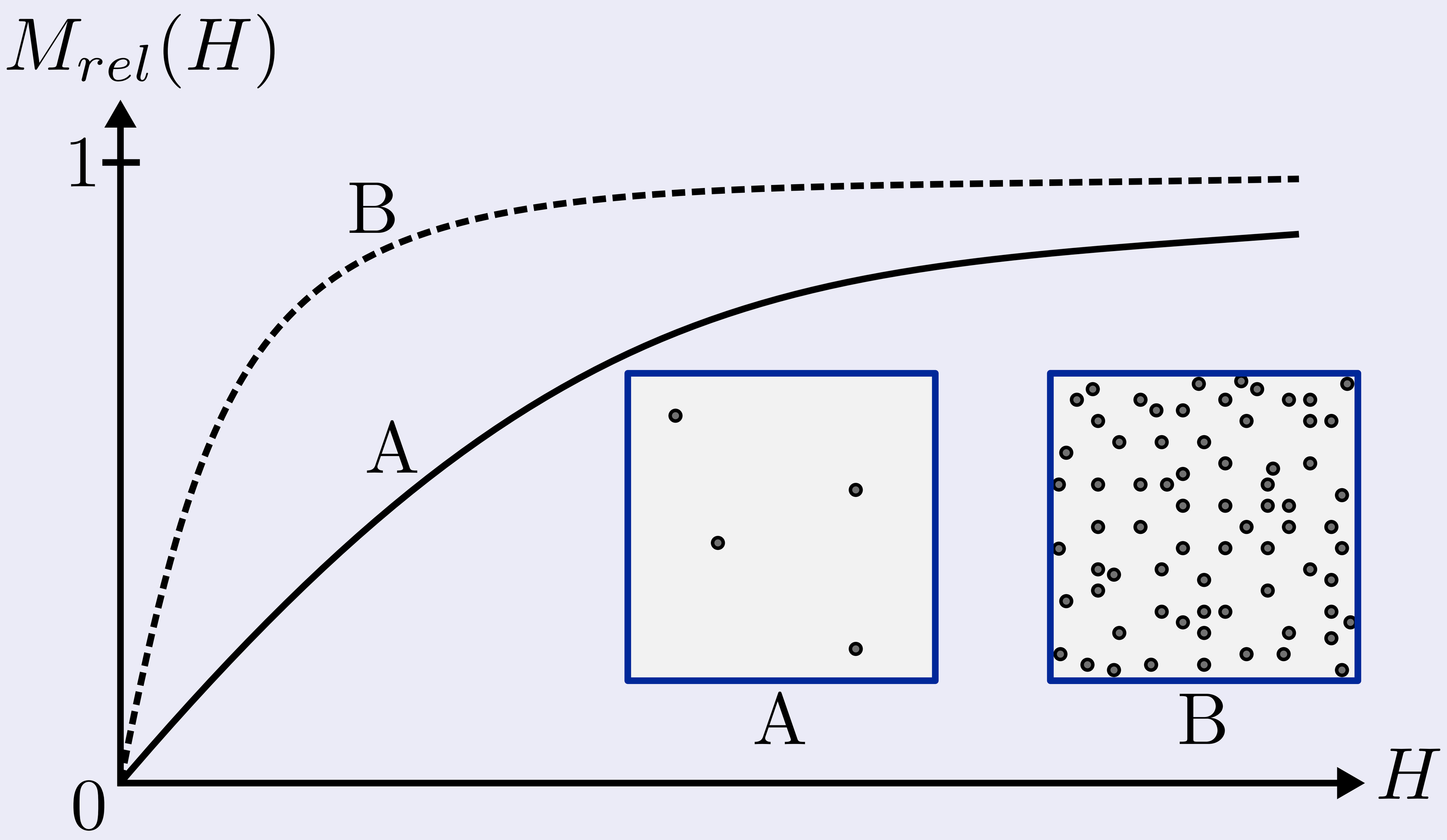

Figure 1: Schematic illustration of relative magnetization curves of two ferrofluids with particles based on the same magnetic cores with diameter d. **Case A**: a very *dilute* ferrofluid (low particle concentration). **Case B**: a quite *dense* ferrofluid (high concentration). Curve A is very near to what the SPM would predict for the diameter d. Curve B strongly deviates from the SPM due to dipole-dipole interaction between the magnetic moments.

MPS Simulation

As a consequence of the nonlinear impact of c on $M(H)$, the n-th higher harmonic amplitude A_n in magnetic particle spectroscopy (**MPS**) is nonlinearly dependent on c, too. For investigation of $A_n(c)$ by means of simulation, we use a second order modified mean-field theory (**MMF2**) [2]. MMF2 incorporates magnetic particle coupling and reflects experimental results best of all models tested in [3].

Within the MPS simulation, a monodisperse ferrofluid made of homogeneously distributed magnetite particles at different iron concentrations c_{Fe} is considered. Using MMF2, the relative magnetization response $M_{rel}(5\,\mathrm{mT}/\mu_0 \sin(\omega_0 t))$ is calculated. Its time derivative is analyzed via discrete FOURIER transformation, leading to $A_n(c_{Fe})$. Evaluation of the *relative* magnetization is equal to analyzing ferrofluids with a *constant* total amount of magnetic substance (shrinking the ferrofluid volume while increasing its concentration).

Figure 2 shows the result of the simulation. To stress the most important outcome, all A_n are normalized to A_3: the ratio A_{n_1}/A_{n_2} changes with varying c_{Fe}. In general, the A_n severely increase due to concentration enhancements. Harmonics with higher n are affected more. As expected, in terms of the SPM the A_n are independent on concentration changes.

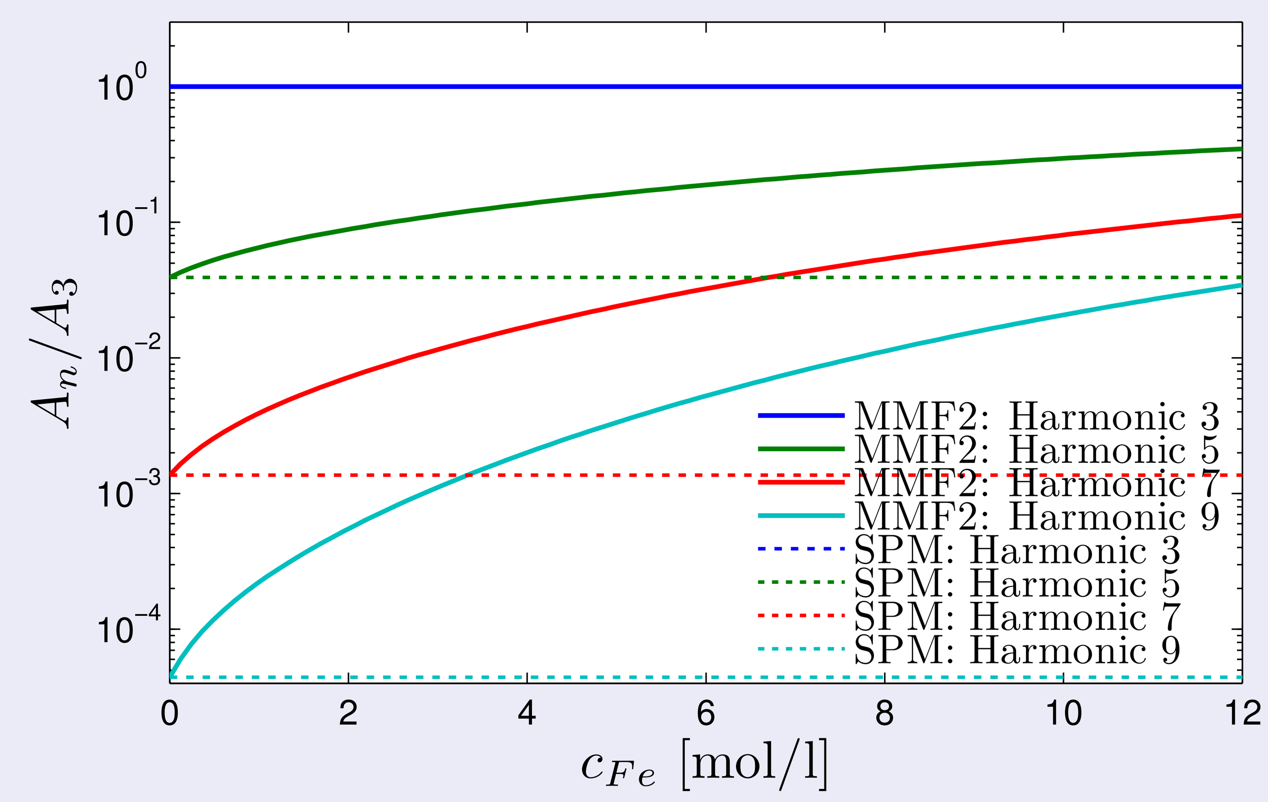

Figure 2: n-th higher harmonic amplitude A_n normalized to A_3 in dependence on iron concentration c_{Fe}, obtained by simulation of the relative magnetization response of a ferrofluid, based on MMF2 theory. The MPS simulation was performed for magnetic particles (saturation magnetization: 480000 A/m; magnetic core diameter: 15 nm) and 5 mT excitation field amplitude.

In biological applications of MPI, strong spatial particle density variations will not be uncommon. An example – leading to iron concentrations of $0.2 - 5$ mol/l in spots – is the agglomeration of particles in cells [4].

MPS Experiment

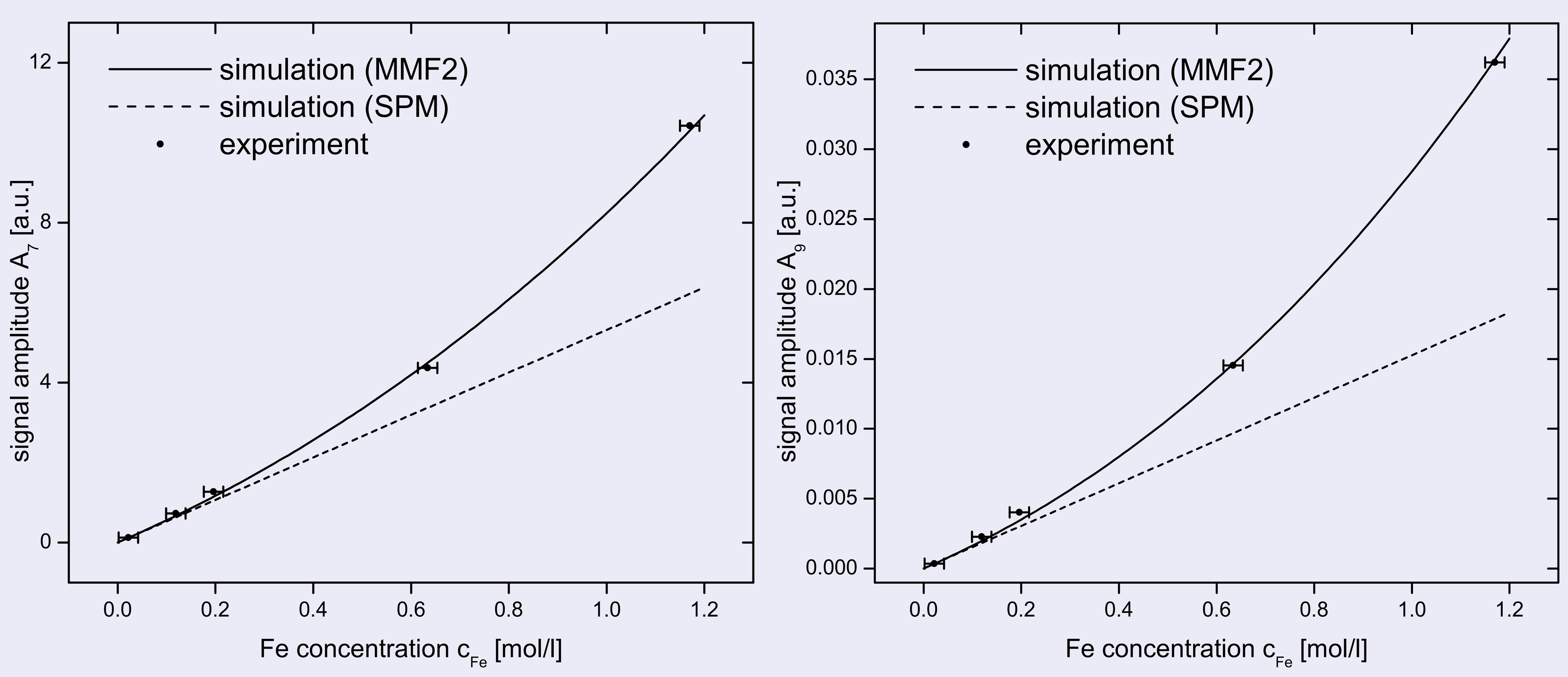

Figure 3: Experimentally gained amplitudes of the 7th (left) and 9th (right) harmonic of the MPS signal of five ferrofluid samples exhibiting different iron concentration c_{Fe}; in comparison with MMF2 and SPM simulations.

$A_7(c)$ and $A_9(c)$ were investigated in an MPS experiment, for one ferrofluid at five different iron concentrations c_{Fe}. The volume of the ferrofluid was kept constant, so that the total amount of substance linearly changes with c_{Fe}. Harmonical excitation was done with 8 mT amplitude. The detected response was averaged 128 times before evaluation. We used a custom ferrofluid made of monodisperse magnetite particles ($d = 8.5$ nm). That allowed us to simulate the experimental setup. This was done using MMF2 and SPM. The result of the simulations was correlated to the experiment by linear scaling of MMF2 data to experimental data.

The experimental and simulated data is shown in Figure 3. The nonlinear $A_n(c)$ dependency could be verified. Furthermore, MMF2 is providing a proper description of the magnetization response of dense ferrofluids. The difference between the SPM and MMF2 curves visualizes the inadequacy of the SPM.

Conclusion

It was shown that changes of the higher harmonic amplitudes in MPI are not linearly related to concentration changes, as misleadingly suggested by the SPM. Therefore, current *linear* image reconstruction schemes are not qualified for quantitative reconstruction of the spatial particle concentration distribution.

For a theoretical description and simulations of the MPI signal, MMF2 seems to be the first choice.

References

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