Non-linear Concentration Effects in Magnetic Particle Imaging

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Introduction
The single particle model (SPM) is a well-established ferrofluid magnetization theory in the field of MPI [1,2]. It describes non-interacting magnetic particles, i.e., particle concentrations c close to 0. In a real ferrofluid, particles interact magnetically. The smaller the distance between single magnetic moments, the stronger their interaction. Hence, the magnetization curve $M(H)$ nonlinearly depends on $c$: especially for increasing $c$ magnetization saturates at lower H. For tomographic purposes, the quantity $c$ has to be reconstructed from the MPI signal. Hence, the quality of the theory describing the impact of concentration $c$ on the magnetization curve is essential for image reconstruction. Since the effect described is ignored by the SPM a different model is required to properly fit data.

Simulations
For our simulations, we use a second order modified mean-field theory (MMF2) [3]. MMF2 incorporates magnetic particle coupling and reflects experimental results best of all models tested in [4]. A mono-disperse ferrofluid made of homogeneously distributed magnetic particles at different iron concentrations $c_{Fe}$ is being considered. Using MMF2, the relative magnetization response $M_d (5mT / p_0 \sin(\phi))$ is calculated. Its time derivative is analyzed via discrete Fourier transformation, leading to amplitudes of higher harmonics $A_n (c_{Fe})$.

As a consequence of the nonlinear impact of $c$ on $M(H)$, the n-th harmonic amplitude $A_n$ in magnetic particle spectroscopy is nonlinearly dependent on $c$, too. As a function of $c_{Fe}$ figure 1 shows the relative magnetization of a sample while figure 2 displays the magnitude of the higher harmonics normalized to the 3rd harmonic are shown. In general, the ratio $A_n / A_3$ severely increase due to concentration enhancements. Harmonics with higher $n$ are affected more. The SPM is, as expected, not able to exhibit this behavior.

To investigate the effect on a 1D image reconstruction, both the currently used techniques, the frequency mixing [5] as well as the drive field [1] were investigated. In order to do reconstructions using the SPM one has to assume an effective particle diameter $d_{eff}$ that compensates for neglecting nonlinear concentration effects in the model. An optimal $d_{eff}$ was obtained by fitting the magnetization curve of SPM to the MMF2 data at a given concentration.

For both imaging techniques the reconstruction quality depends heavily on the choice of the correct $d_{eff}$. As an example figure 3 shows the effects of a wrong estimation of $d_{eff}$ in case of the drive field method.

Experiments
$A_7 (c)$ and $A_{d7} (c)$ were investigated in an MPS experiment for one ferrofluid of mono-disperse magnetite particles ($d = 8.5 \text{ nm}$) at five different iron concentrations $c_{Fe}$ to validate our simulations. The volume of the ferrofluid was kept constant, so that the total amount of experiment by linear scaling of MMF2 data to experimental data. The experimental and simulated data is shown in figure 4. The nonlinear $A_{d7}(c)$ dependency could be verified. Furthermore, MMF2 is providing a proper description of the magnetization response of dense ferrofluids.

Conclusions
Higher harmonic amplitudes in MPI are not linearly related to concentration changes, as suggested by the SPM. Therefore, current linear image reconstruction schemes will fail in properly reconstructing spatial particle concentration distributions containing areas with dense ferrofluids. Those will not be uncommon in biological applications of MPI, e.g. the agglomeration of particles in cells – leading to iron concentrations of $0.2 \sim 5 \text{ mol/l}$ in localized areas [6]. Thus, incorporating this non-linear behavior in imaging reconstruction schemes is mandatory. The experimental results suggest that using MMF2 is capable of correctly describing the non-linear effects on the higher harmonics in the MPI signal. The difference between the SPM and MMF2 curves visualizes the inadequacy of the SPM. Therefore it should be used instead of SPM for dense ferrofluids.

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References

Figure 1: (top) Normalized magnetization curves for different concentrations $c$, calculated using MMF2, in comparison with SPM.
Figure 2: (bottom) Normalized signal amplitudes of a spectroscopic MPI experiment in dependence of $c$. Signal obtained by MMF2 and SPM in comparison.
Figure 4: Experimentally gained amplitudes of the $7^{th}$ harmonic of the signal of five ferrofluid samples exhibiting different iron concentration $c_{Fe}$, in comparison with MMF2 and SPM simulations.