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How MNP respond to dual-frequency magnetic excitation in viscous media: an in silico study

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Abstract

Magnetic nanoparticles (MNP) enable new biomedical applications as imaging tracers, heating agents or biosensors due to their unique relaxation mechanism in alternating magnetic fields. For assessing MNP suitable for such applications, magnetic particle spectroscopy (MPS) offers a reliable method, dual-frequency excitation adding sensitivity. Biomedical applications, however, rely on MNP use in physiological environments (blood, tissue, etc.) of various viscosities, which could strongly alter the MNP relaxation behavior. In this work, we present our preliminary results of varying viscosity on the relaxation of MNP during dual-frequency MPS, studied with micromagnetic dynamic magnetization simulation.

I. Introduction

Magnetic nanoparticles (MNP) are of great interest for biomedical applications in diagnostics with magnetic particle imaging, hyperthermia treatment, and MPS-based biosensing. With special regard to biosensing, dual-frequency magnetic particle spectroscopy, known as frequency mixing magnetic detection (FMMD), offers a more sensitive method by introducing frequency mixing terms in the MNP's signal response. These intermodulation frequencies allow additional information recovery from the MNP and are uniquely characterizing their properties. The above applications of MNP rely on the magnetic relaxation mechanism of MNP to an alternating magnetic field that has been extensively described by stochastic dynamic modeling in the past decade, enabling sophisticated micromagnetic Monte-Carlo-based dynamic micromagnetic simulations (MCS) to study the effects of individual parameters independently [1]: i.e. being able to simulate Brownian relaxation and thereby viscosity effect on MNP relaxation, which Langevin modelling in thermal equilibrium cannot [2]. Clinical applications rely on the use in physiological environments (i.e. blood, tissue, cells). The relaxation mechanism is strongly influenced by the surrounding medium, expressed by the viscosity [1]. Here, we present preliminary results on simulating the influence of viscosity on the FMMD signal of MNP.

II. Methods and materials

We used MCS to predict the magnetic response of magnetite MNP ($M_s = 476\ \mathrm{kAm^{-1}}$, $K_u = 11\ \mathrm{kJ{\cdot}m^{-3}}$, $T = 298\ \mathrm{K}$)

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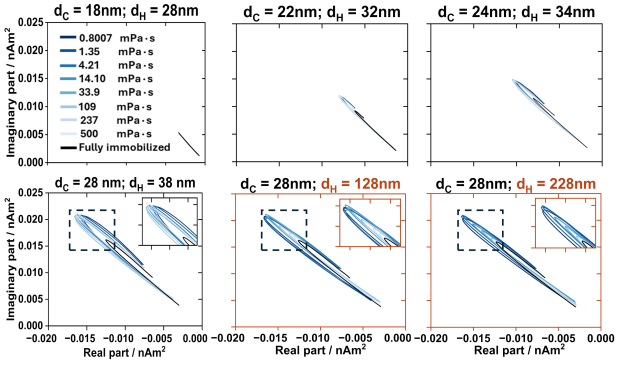

Figure 1: Real-vs-imaginary part of the complex FMMD signal of mixing frequency $A_{f_1+2f_2}$ for increasing viscosity until the MNP are fully immobilized. Orange borders indicate higher shell thickness for $d_c = 28nm$. Insets show zoom on turning points.

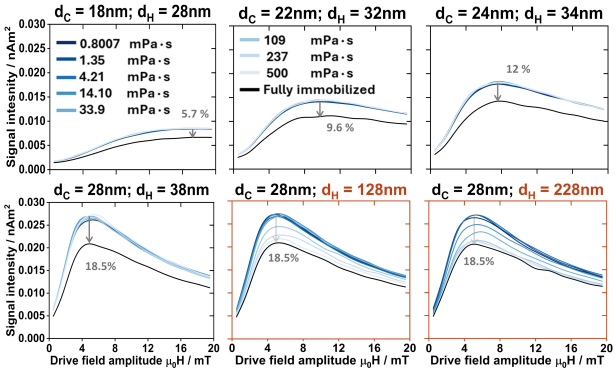

Figure 2: FMMD signal intensity of mixing frequency term $A_{f_1+2f_2}$ for increasing viscosity until the MNP are fully immobilized. Gray arrows and values show the reduction in signal intensity upon full immobilization of particles at peak intensity. Orange borders indicate higher shell thickness for $d_c = 28nm$.

to a dual frequency magnetic excitation with the field [2]

$$H(t) = H_1 \sin\left(2\pi f_1 t\right) + H_2 \sin\left(2\pi f_2 t\right), \qquad (1)$$

where $f_1 = 40.5$ kHz, $H_1 = 1.29$ mT/μ_0, and $f_2 = 63$ Hz were constant for varying drive fields H_2 varied from 0.5 to 19.5 mT/μ_0 in steps of 1.5 mT/μ_0. We assume uniaxial excitation field applied in the z-direction (arbitrarily chosen) as well as uniaxial anisotropy. We simulated the M(H) curves (dynamic magnetization loops) for core sizes of $d_C = (18, 22, 24, 28)$ nm with size distribution width $\sigma = 0.05$, and hydrodynamic size of $d_H = d_C + 2 \cdot d_S$. Shell thickness was $d_S = 5$ nm for all cores and additionally $d_S = (50, 100)$ nm for $d_C = 28$nm. We probed for increasing viscosity, starting from water viscosity ($\eta = 0.8007$ mPa s) to 99% Glycerol concentration ($\eta = 500$ mPa s), until full immobilization.

III. Results and discussion

Figure 1 shows the FMMD signal of the mixing frequency term, $A_{f_1+2f_2}$, in the complex plane for all MNP. Obviously, the core size strongly dominates the FMMD signal, while the effect of viscosity is only visible for $d_H > 100$ nm (s. lower row in Figure 1). An exception are fully immobilized MNP, whose signal is reduced significantly by almost 20% for all samples, independent of d_H. A closer look at larger MNP ($d_C = (24; 28)$ nm) predicts a slight increase in the imaginary parts with increasing viscosity. The positions of the turning-points (s. inset in Figure 1), however, drop visibly only for larger shell thicknesses for $d_C = 28$ nm at the following viscosities: $\eta^*(d_S = 50\,\text{nm}) \geq 109$ mPa s (85% Glycerol), and $\eta^*(d_S = 100\,\text{nm}) \geq 14.4$ mPa s (65% Glycerol). The same effects are also observed in Figure 2 for the signal intensity of $A_{f_1+2f_2}$, showing (a) strong size-dependence and a maximum signal decrease by 5.7 % (18 nm) to 18.5 % (28 nm) for fully immobilized MNP; (b) gradually decrease in signal intensity at the same viscosities, $\eta^*(d_S)$, as observed above, and (c) maximally decreased signal (at level of MNP immobilized) for viscosities $\eta^*(d_S = 50\,\text{nm}) \geq 500$ mPa s and $\eta^*(d_S = 100\,\text{nm}) \geq 109$ mPa s.

Increasing viscosity yields larger Brownian relaxation times [3], resulting in a delayed magnetic response and an increase in the imaginary parts' contribution to the FMMD signal, as observed in Figure 1. As we purposefully simulated particles of low hydrodynamic sizes ($d_H < 40$ nm), the effect of viscosity on these MNP is expected to be small [3]. Still being able to detect this change demonstrates the high sensitivity of FMMD (cf. Figure 1). However, larger shell thicknesses ($d_H > 100$ nm) are increasingly affected by viscosity, gradually decreasing in FMMD (complex) signal and intensity with increasing viscosity until reaching maximum reduction for fully immobilized particles, as similarly observed for the heating performance of immobilized MNP [4].

IV. Conclusion

Our preliminary study suggests a dominant effect from core sizes over viscosity, which has minimal effect on MNP of small hydrodynamic size ($d_H < 40$ nm), but which is quantifiable for larger core sizes, $d_C \geq 24$ nm by FMMD. Larger $d_H > 100$ nm allows for gradual reduction of the FMMD signal, maximally reduced for fully immobilized MNP with blocked Brownian relaxation (drop of 18.5% at $d_C = 28$ nm). This suggests that viscosity could play a significant role for FMMD signal generation. We plan to compare our preliminary results to experimental data and to simulate larger viscosities to further quantify the effect of viscosity on Brownian relaxation.

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Author's statement

Authors state no conflict of interest.

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