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Effect of viscosity on harmonic signals from magnetic fluid

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ABSTRACT

We explored the effect of viscosity on harmonic signals from a magnetic fluid. Using a numerical simulation that accounts for both the Brownian and Néel processes, we clarified how the magnetization mechanism is affected by viscosity. When the excitation field varies much slower than the Brownian relaxation time, magnetization can be described by the Langevin function. On the other hand, for the case when the excitation field varies much faster than the Brownian relaxation time, but much slower than the Néel relaxation time, the easy axes of the magnetic nanoparticles (MNPs) turn to some extent toward the direction of the excitation field in an equilibrium state. This alignment of the easy axes of MNPs caused by the AC field becomes more significant with the increase of the AC field strength. Consequently, the magnetization is different from the Langevin function even though Néel relaxation time is faster than time period of the external frequency. It is necessary to consider these results when we use harmonic signals from a magnetic fluid in a high-viscosity medium.

1. Introduction

Magnetic nanoparticles (MNPs) in solution, i.e., magnetic fluids, have been widely studied for biomedical applications such as separation of biological targets, immunoassays, drug delivery, hyperthermia, and magnetic particle imaging (MPI) [1]. MPI presents a new modality for imaging the spatial distribution of MNPs, especially for in-vivo diagnostics [2]. Thus far, several methods, such as harmonic-space MPI [2–4], narrowband MPI, [5] and *x*-space MPI [6,7], have been introduced to reconstruct the image of the spatial distribution of the MNPs in MPI. In harmonic-space MPI, harmonic signals generated by the nonlinear magnetization of MNPs exposed to both an AC excitation field and a DC gradient field are detected to image the spatial distribution of MNPs.

In order to characterize or optimize the MNPs for MPI application, harmonic magnetization signals from MNPs have been studied. In Ref. [8], the Langevin function, which describes the static magnetization of superparamagnetic MNPs, was used to evaluate the harmonic signals. In order to take the finite relaxation time into account, a modified Langevin function was used in Refs. [9,10]. However, this modified Langevin function has not been theoretically established. On the other hand, in Ref. [11], the stochastic Landau–Lifshitz–Gilbert (LLG) equation, which takes into account thermal fluctuations, was used to evaluate the harmonic signals from MNPs for MPI application. While the stochastic LLG equation describes the dynamic behavior of the magnetization of MNPs, the numerical simulation in Ref. [11] was restricted to the case of immobilized MNPs.

It is well recognized that magnetization in a magnetic fluid occurs via Néel and Brownian processes. Since the Brownian relaxation time is proportional to the viscosity of the surrounding medium and the viscosity may change in practical applications, it is important to investigate the effect of viscosity on the harmonic signals from a magnetic fluid.

In this study, we investigate the dynamic behavior of MNPs by considering both the Néel and Brownian relaxations. We first show that the measured third harmonic signals from two magnetic fluid samples with different viscosities exhibit different properties. Then, the effect of viscosity on the mechanism of magnetization in the magnetic fluid is clarified with a numerical simulation. Finally, based on the numerical simulation results, we obtain some empirical equations for the magnetization of MNPs.

2. Experimental

2.1. Materials and methods

In the experiment, commercial MNPs called Resovist (FUJIFILM RI Pharma) were used as samples. Resovist is a hydrophilic colloidal solution of $Fe_3O_4/\gamma Fe_2O_3$ nanoparticles coated with carboxydextran, which has a primary core diameter in the range of 5–10 nm. It consists of clusters of elementary particles. The iron

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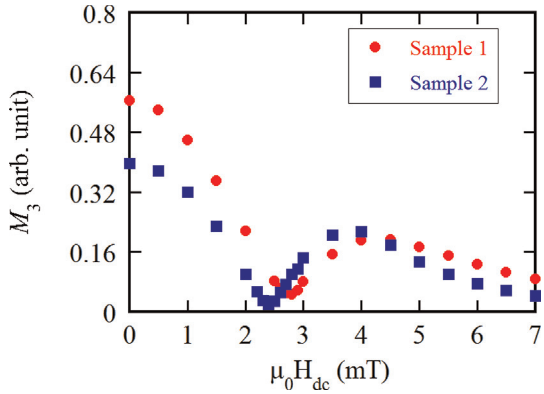

Fig. 1. Experimental results of the third harmonics vs. DC field when an excitation field with $\mu_0 H_{ac}=3$ mT and $f=1$ kHz was applied. Samples 1 and 2 were diluted with pure water and glycerol, respectively.

concentration of the original Resovist magnetic fluid is 27.875 mg (Fe)/ml.

To investigate the effect of viscosity on harmonic signals from a magnetic fluid, we prepared two samples. In sample 1, 60 µl of the Resovist magnetic fluid was diluted in 230 µl of pure water, while in sample 2, 230 µl of glycerol was substituted in place of water. These two samples differ in viscosity.

In the experiment, an external field, $H=H_{dc}+\sqrt{2}H_{ac}\sin(2\pi ft)$, was applied via an excitation coil. The magnetic signal from the MNPs was detected using an inductive pickup-coil gradiometer installed concentrically with the excitation coil. In order to avoid interference from the excitation field, a first-order gradiometer was used.

2.2. Experimental results

In Fig. 1, experimental results of the third harmonic signal vs. DC field are shown. AC excitation field with $\mu_0 H_{ac}=3$ mT and $f=1$ kHz was applied. As shown, the DC field dependence of the third harmonic was different between the two samples, which indicated that viscosity affected the third harmonic signal. Since the DC field dependence of the harmonic signal is directly related to the spatial resolution in MPI, this result implies that spatial resolution is also affected by the viscosity of the magnetic fluid.

We note that the vertical axis, M_3, represents the absolute value of the third harmonic signal. It can be shown from the Langevin function that the third harmonic signal decreases, becomes zero at a certain DC field, and then becomes negative values when the DC field is increased [12]. Therefore, if we plot the absolute value of the third harmonic signal, it shows the sharp minimum at a certain DC field, as shown in Fig. 1.

3. Numerical simulation

3.1. Methods

We performed numerical simulations to study the mechanism causing the difference in third harmonics between samples 1 and 2. When the MNPs can rotate physically, the dynamics of the magnetic moment is determined by a combination of Brownian and Néel processes.

The dynamics of a unit vector along the easy axis $\vec{n}$ is given by the Langevin equation [13]

$$\frac{d\vec{n}}{dt}=\frac{1}{\eta\pi d_h^3}\vec{m}\times\vec{B}\times\vec{n}+\sqrt{\frac{k_BT}{\eta\pi d_h^3}}\vec{\Gamma}\times\vec{n}, \tag{1}$$

where η is the viscosity of the surrounding medium, d_h is the hydrodynamic size of the MNP, $\vec{m}$ is the magnetic moment vector of the MNP, and k_BT is the thermal energy. The random torque, $\vec{\Gamma}$, on the MNP caused by thermal fluctuations satisfies the following equations:

$$\langle\Gamma_i\rangle=0, \tag{2}$$

$$\langle\Gamma_i(t)\Gamma_j(t')\rangle=2\delta_{ij}\delta(t-t'). \tag{3}$$

Here,⟨⟩ represents the average over an ensemble, i and j are Cartesian indices, δ_{ij} is the Kronecker delta function, and δ is the Dirac delta function.

On the other hand, the dynamics of the magnetic moment vector $\vec{m}$ is given by [13]

$$\frac{d\vec{m}}{dt}=\gamma\vec{m}\times\left(\vec{B}_{eff}+\vec{B}_{th}\right)-\gamma\frac{\lambda}{m}\vec{m}\times\left[\vec{m}\times\left(\vec{B}_{eff}+\vec{B}_{th}\right)\right] \tag{4}$$

where γ is the gyromagnetic ratio and λ is a dimensionless damping coefficient. $\vec{B}_{eff}$ is the effective magnetic field given by

$$\vec{B}_{eff}=\vec{B}_{ex}+\vec{B}_{ani}, \tag{5}$$

$$\vec{B}_{ani}=\frac{2KV_c}{m}\frac{\vec{m}\cdot\vec{n}}{m}\vec{n},, \tag{6}$$

where $\vec{B}_{ex}$ is the external field and $\vec{B}_{ani}$ is the magnetic-anisotropy field. $\vec{B}_{th}$, the fluctuating magnetic field due to thermal noise, satisfies the following equations:

$$\langle B_{th,i}(t)\rangle=0, \tag{7}$$

$$\langle B_{th,i}(t)B_{th,j}(t')\rangle=\frac{2\lambda}{1+\lambda^2}\frac{k_BT}{\gamma m}\delta_{ij}\delta(t-t') \tag{8}$$

Eq. (4) is a stochastic LLG equation in which thermal fluctuations are taken into account.

The dynamic behavior of the magnetic moment as well as the easy axis for a single MNP can be calculated by solving Eqs. (1) and (4) simultaneously. In the simulation, Eqs. (1) and (4) were discretized with respect to time t. The discretization interval Δt was set to $0.01\tau_{N0}$, and the value $\lambda=0.1$ was used. Here, $\tau_{N0}=m/(2\gamma\lambda E_B)=10^{-9}$ is the characteristic Néel relaxation time, $E_B=K\pi d_c^3/6$ is the anisotropy energy barrier, K is the anisotropy constant, and d_c is the core size of the MNP. The dynamics of the magnetic moment vector $\vec{m}$ and unit vector along the easy axis $\vec{n}$ were calculated for $N=7168$ MNPs. Consequently, the average value $\langle\vec{m}\rangle$ over the ensemble was obtained. The numerical simulation was carried out until an equilibrium magnetization was obtained. The harmonic spectrum of the magnetization was obtained by performing a Fourier transform on this equilibrium magnetization.

For simplicity, we assumed that all the MNPs have the same values of m and E_B in the simulation, although the real Resovist sample has a size distribution. In Ref. [11], it was shown that Resovist MNPs with 15 nm $< d_c <$ 40 nm size distribution exhibited a rich harmonic spectrum. Therefore, we set $d_c=28$ nm in the simulation. Further, $M_s=360$ kA/m, $K=4$ kJ/m^3, and $t_{co}=7$ nm were used, as described in Ref. [11]. Here, $t_{co}=(d_h-d_c)/2$ is the thickness of the coating material. The Néel relaxation time was calculated to