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Individual observation of Néel and Brownian relaxations in magnetic nanoparticles

S. Ota[1,*] · Y. Takemura[2]

[1]Department of Electrical and Electronic Engineering, Shizuoka University, Hamamatsu, Japan
[2]Department of Electrical and Computer Engineering, Yokohama National University, Yokohama, Japan
*Corresponding author, email: ota.s@shizuoka.ac.jp

Abstract

Magnetic relaxation divided into the Néel and Brownian regimes determines the magnetization dynamics. To develop the signal intensity and resolution of magnetic particle imaging, understanding the magnetization dynamics was required. In this study, the Néel and Brownian relaxations were individually evaluated. The two-step magnetization response of magnetic nanoparticles dispersed in a fluid as the Brownian regime occurred after the Néel regime was observed by applying a fast responding pulse field. To clarify the magnetic relaxation in detail, it is necessary to individually observe the Néel and Brownian relaxations in the superposition system. By fitting the theoretical calculation to the measured time evolution of the magnetization response, we isolated Néel and Brownian relaxations from the experimentally observed superposition relaxation system. Moreover, the effect of dipole interactions on Néel and Brownian relaxation were confirmed by measuring the dependence of magnetization responses on the particle concentration in a magnetic fluid.

I Introduction

To develop the sensitivity and resolution of magnetic particle imaging (MPI) [1], it is necessary to understand the magnetization dynamics determined by the magnetic relaxation. The harmonics of magnetization signal are detected in MPI, which is associated with the non-linear response of the magnetization.

The Néel and Brownian relaxations as the relaxations of the magnetization and the easy axis were occurred along the effective relaxation time τ_{eff}, given by $1/\tau_{\text{eff}} = 1/\tau_N N + 1/\tau_B$, where τ_N and τ_B were the Néel and Brownian relaxation times [2]. This indicates the dominance of one relaxation with the relaxation time shorter than other. On the other hand, the limitation of the model using the effective relaxation time was indicated [3]. We observed the superposition of the Néel and Brownian relaxations by applying AC magnetic field and pulse field [4,5]. The superposition of Néel and Brownian relaxations was also shown by measurement, calculation, and numerical simulation in conventional studies [6–8].

In this study, we experimentally observed the individual relaxation processes in a superposition system of Néel and Brownian relaxations, which were not empirically evaluated [9]. This is because the magnetization rotation derived from Néel relaxation is too fast to allow sufficient measurement of the magnetic flux alteration of MNPs, particularly in the case of superparamagnetic behavior.

II Material and methods

A water-based Fe_3O_4 nanoparticle, commercially distributed as M-300 by Sigma Hi-chemical Inc., and coated with α-olefin sulphonic acid sodium, was measured. The nanoparticle core and hydrodynamic diameters were determined to be a mean of 11 nm and 44 nm measured

by transmission electron microscopy and dynamic light scattering, respectively. The measured samples were prepared by dispersing the particles in diluted water in the concentrations adjusted to 1.24, 2.47, 4.95, and 9.89 % v/v.

Figure 1a shows the effective exciting pulse field, whose maximum intensity and rise time were 0.38 kA/m and 18 ns, respectively. To detect a magnetization flux, we used two different types of pick-up coil that had small and large numbers of turns, with low and high inductance, respectively.

The magnetization response to the pulse field is given by

$$M_{\mathrm{eff}}(t) = M_N(t) + M_B(t), \tag{1}$$

$$M_{\mathrm{R}}(t) = M_{\mathrm{R,max}}\left\{1 - \exp\left(-\frac{t}{\tau_r}\right)\right\}, \tag{2}$$

where $M_{\mathrm{eff}}(t)$ is the time depending effective magnetization, which is the superposition of the magnetizations depending on the Néel and Brownian relaxations, $M_N(t)$ and $M_B(t)$, whose maximum values, $M_{\mathrm{N,max}}$ and $M_{\mathrm{B,max}}$, are substituted into the general expression for the magnetization $M_R(t)$ [5]. τ_N and τ_B are substituted into the general expression for the relaxation time τ_R, in terms of the response associated with Néel and Brownian relaxations, respectively.

In addition, the magnetization response was calculated in consideration of the size distribution of MNPs with respect to core and hydrodynamic diameters, d_C and d_H, which are substituted into the general term for the diameter d_R,

$$M_R(t) = \sum_{i=1}^{n} F(d_{R,i}) M_{R,i}(t) / \sum_{i=1}^{n} F(d_{R,i}), \tag{3}$$

$$F(d_{R,i}) = \frac{1}{d_{\mathrm{R,std}}\sqrt{2\pi}} \exp\left(-\frac{d_{R,i} - d_{\mathrm{R,ave}}}{2d_{\mathrm{R,std}}^2}\right) \tag{4}$$

where the n is the number of particles, and i is the index for each particle. $F(d_R, i)$ are the possibility density functions with respect to the Gaussian distribution. $d_{\mathrm{R,ave}}$ and $d_{\mathrm{R,std}}$ are the average value and standard deviation of d_R.

III Results and discussion

Figure 1b shows the measured magnetization M_{exp} in a nanoparticle concentration of 1.24 % v/v. The magnetization process was first promoted by Néel rotation, then further enhanced by Brownian rotation. In the time range shorter than 18 ns, because the magnetization was enhanced along with increasing field intensity, the magnetization process was not fitted by the theoretical model in Néel regime. Using the fitting curves obtained by eqs. (1–3), the magnetization responses were divided into the

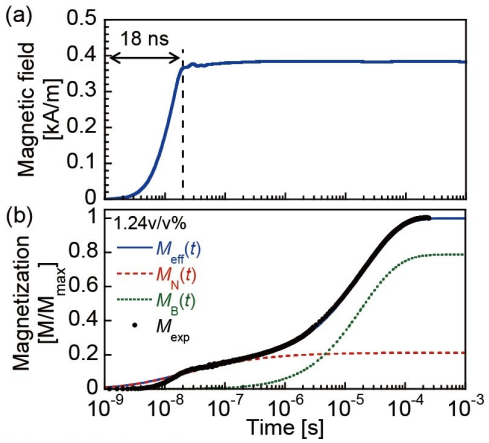

Figure 1: (a) Time evolution of the applied pulse field. (b) Calculated magnetization of the effective response, $M_{\mathrm{eff}}(t)$, in the Néel regime, $M_{\mathrm{N}}(t)$, and Brownian regime, $M_{\mathrm{B}}(t)$, and the experimental magnetization response, $M_{\mathrm{exp}}(t)$.

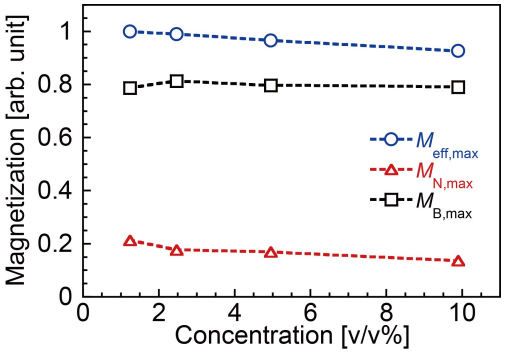

Figure 2: Concentration dependence of the calculated maximal values for the effective magnetization, $M_{\mathrm{eff,max}}$, magnetization in the Néel regime, $M_{\mathrm{N,max}}$, and magnetization in the Brownian regime, M_{Bmax}.

individual responses belonging to Néel and Brownian regimes. The estimated core and hydrodynamic diameters were 11.9±2 nm and 40±11 nm (mean ± SD), which was in good agreement with the measured diameters. With respect to M_{exp} in the concentration of 2.47, 4.95, and 9.89 % v/v, $M_N(t)$ and $M_B(t)$ was also calculated using eqs. (1–3).

Figure 2 shows the maximal values in $M_{\mathrm{eff}}(t)$, $M_N(t)$, and $M_B(t)$, $M_{\mathrm{eff,max}}$, $M_{\mathrm{N,max}}$, and $M_{\mathrm{B,max}}$, depended on the particle concentration, respectively. With increasing concentration, $M_{\mathrm{eff,max}}$ and $M_{\mathrm{N,max}}$ were decreased, and $M_{\mathrm{B,max}}$ was constant. It is indicated that the magnetization in the Néel regime was decreased due to the dipole interaction because of the short distance among particles in the high concentration, and the magnetization in the Brownian regime was not affected by the dipole interaction. With respect to the concentration dependence of dipole interactions observed by measurements, the dipolar field increased with increasing concentration, which

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inhibited the magnetization [10]. On the other hand, Ovejero *et al.* suggested that intra-aggregate interactions such as dipole interactions over short interparticle distances reduce magnetization whereas inter-aggregate interactions over long interparticle distances enhance magnetization [11]. The dipole interaction in the Néel regime was associated with the short interparticle distances in aggregation in hydrodynamic condition.

The ratio of $M_{\mathrm{B,max}}$ to $M_{\mathrm{N,max}}$, $M_{\mathrm{B,max}}/M_{\mathrm{N,max}}$, in 1.24, 2.47, 4.95, and 9.89 % v/v estimated from the fitting curves were 3.7, 4.6, 4.7, and 5.8, respectively. In previous study, we show that the harmonic signal of MNPs of large anisotropy in liquid was significantly larger than those in solid because of the Brownian rotation in liquid [12]. Thus, the dominance of the Brownian rotation in the magnetization dynamics, $M_{\mathrm{B,max}}/M_{\mathrm{N,max}}$, influenced by the effective anisotropy was associated with the harmonic intensity.

IV Conclusions

We experimentally observed the superposition of the Néel and Brownian relaxations by applying a low-intensity pulse field with a rise time shorter than the Néel relaxation time. The magnetization responses in Néel and Brownian regimes were individually isolated from the superposition system using a theoretical fitting procedure that considered size distribution. Moreover, the dominance of the magnetization response depended on the particle concentration as the effect of the dipole interaction was evaluated. By clarifying the magnetization dynamics associated with the magnetic relaxation, the harmonic signal in MPI derived from the non-linear response of MNPs may be improved.

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References

[1] B. Gleich and J. Weizenecker. Tomographic imaging using the nonlinear response of magnetic particles. Nature, 435, 1217-1217, 2005.

[2] R.E. Rosensweig, Heating magnetic fluid with alternating magnetic field. J. Magn. Magn. Mater., 5, 370–374, 2002.

[3] M. I. Shliomis and V. I. Stepanov, Theory of the dynamic susceptibility of magnetic fluids, in relaxation phenomena in condensed matter, Adv. Chem. Phys., 87, 1-30, 1994

[4] S. Ota, T. Yamada, and Y. Takemura, Dipole-dipole interaction and its concentration dependence of magnetic fluid evaluated by alternating current hysteresis measurement, J. Appl. Phys., 117, 17D713, 2015.

[5] Suko Bagus Trisnanto, Satoshi Ota, and Yasushi Takemura, Two-step relaxation process of colloidal magnetic nanoclusters under pulsed fields, Appl. Phys. Express, 11, 075001, 2018.

[6] R. Kötitz, W. Weitschies, L. Trahms, W. Brewer, W. Semmler, determination of the binding reaction between avidin and biotin by relaxation measurements of magnetic nanoparticles. J. Magn. Magn. Mater., 194, 62-68, 1999.

[7] R. Hergt, R. Hiergeist, I. Hilger, W. A. Kaiser, Y. Lapatnikov, S. Margel, U. Richter, Maghemite nanoparticles with very high AC-losses for application in RF-magnetic hyperthermia. J. Magn. Magn. Mater., 270, 345-357, 2004.

[8] H. Mamiya and B. Jeyadevan, Hyperthermic Effects of Dissipative structures of magnetic nanoparticles in large alternating magnetic fields. Sci. Rep., 1, 157, 2011.

[9] S. Ota and Y. Takemura, Characterization of Néel and Brownian relaxations isolated from complex dynamics influenced by dipole interactions in magn etic nanoparticles. J. Phys. Chem. C, 123, 28859-28866, 2019.

[10] S. Dutz and R. Hergt, The role of interactions in systems of single domain ferrimagnetic iron oxide nanoparticles. J. Nano- Elect. Phys., 4, 02010, 2012.

[11] J. G. Ovejero, D. Cabrera, J. Carrey, T. Valdivielso, G. Salas, F. J. Teran, Effects of inter- and intra-aggregate magnetic dipolar interactions on the magnetic heating efficiency of iron oxide nanoparticles. Phys. Chem. Chem. Phys,. 18, 10954-10963, 2016.

[12] S. Ota, Y. Matsugi, T. Nakamura, R. Takeda, Y. Takemura, I. Kato, S. Nohara, T. Sasayama, T. Yoshida, and K. Enpuku, Effects of size and anisotropy of magnetic nanoparticles associated with dynamics of easy axis for magnetic particle imaging, J. Magn. Magn. Mater., 474, 311–318, 2019.

