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The Effect of Nonmagnetic Particles on the Magnetic Interactions and Microstructure in Magnetic Inks

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The magnetic properties and performance of magnetic inks are strongly dependent on the interparticle interactions and the quality of the particle dispersion. In this study, we investigated the effect of diluting magnetic ink by replacing magnetic Co-γ-Fe_2O_3 particles with nonmagnetic α-Fe_2O_3 particles (0~50 wt%). The magnetic interactions and microstructure of the wet inks were characterized using three methods: transverse susceptibility (χ_t), rheological measurements (yield stress, τ_y), and complex magnetic susceptibility. As the concentration of nonmagnetic particles increased, the DC magnetic field strength (H_{peak}) corresponding to the maximum transverse susceptibility shifted to lower fields, indicating easier orientation of the magnetic particles. Rheological measurements showed that the yield stress decreased linearly with the magnetic particle volume fraction. Furthermore, frequency-dependent susceptibility measurements showed that the low-frequency susceptibility (χ'_0) and magnetic relaxation time (τ) both increased with dilution. These results consistently demonstrate that the addition of nonmagnetic particles reduces interparticle magnetic interactions, improving the dispersion quality by breaking down particle agglomerates and increasing the population of single particles.

Keywords : magnetic susceptibility, yield stress, magnetic field strength, magnetic ink, nonmagnetic particles

자성 잉크에서 비자성 입자가 자기적 상호작용과 미세구조에 미치는 영향

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자성 잉크의 자기적 성질과 성능은 입자 간 상호작용과 분산 품질에 크게 좌우된다. 본 연구에서는 자성 Co-γ-Fe_2O_3 입자를 비자성 α-Fe_2O_3 입자(0~50 wt%)로 치환하여 자성 잉크를 희석했을 때의 효과를 조사하였다. 자성 잉크의 자기적 상호작용과 미세구조를 횡자화율(χ_t), 레올로지 측정(항복응력, τ_y), 복소 자기 감수율의 세 가지 방법으로 관찰하였다. 비자성 입자 농도가 증가함에 따라 최대 횡자화율에 대응하는 DC 자기장 세기(H_{peak})가 더 낮은 값으로 이동했으며, 이는 자성 입자의 배향이 더 용이해졌음을 나타낸다. 레올로지 측정 결과, 항복응력은 자성 입자 체적분율에 따라 선형적으로 감소하였다. 더 나아가, 주파수 의존 감수율 측정에서 저주파 감수율(χ'_0)과 자기 이완 시간(τ)이 희석과 함께 모두 증가함을 보였다. 이러한 결과는 일관되게, 비자성 입자의 첨가가 입자 간 자기적 상호작용을 약화시켜 응집체를 분해하고 단일 입자의 비율을 증가시킴으로써 분산 품질을 향상시킨다는 것을 보여준다.

주제어 : 자기 감수율, 항복응력, 자기장 세기, 자성 잉크, 비자성 입자

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I. Introduction

Magnetic recording media remains a cornerstone of high-capacity data storage technology [1-5]. The continuous demand for higher areal density storage necessitates a persistent improvement in media performance, which is critically dependent on maximizing the signal-to-noise ratio (SNR). In particulate recording media, a primary source of noise arises from the microstructural non-uniformity of the magnetic coating, which is directly determined by the quality of the particle dispersion in the liquid magnetic ink.

Magnetic inks are complex colloidal suspensions, typically composed of acicular (needle-like) magnetic particles (e.g., Co-γ-Fe_2O_3) dispersed in a polymer resin and solvent system. Due to their permanent magnetic moments, these particles are subject to strong, long-range magnetic dipole-dipole interactions, in addition to ubiquitous van der Waals forces. These powerful attractive forces inevitably lead to the formation of particle agglomerates and flocculated, network-like structures within the wet ink [6-8].

This agglomeration is highly detrimental to recording performance. When a poorly dispersed ink is coated and dried, the resulting film exhibits increased surface roughness [9] and significant magnetic inhomogeneity. Large particle clusters cannot be effectively and uniformly oriented by the recording head's magnetic field, leading to increased transition noise, reduced signal output, and ultimately, poor recording performance [10]. Therefore, achieving a stable and uniform dispersion of individual magnetic particles in the wet ink state is a paramount challenge.

Various strategies are employed to overcome these interparticle attractions. The most common approach involves the adsorption of polymers (binders or resins) and surfactants onto the particle surfaces, which provide steric or electrostatic stabilization to counteract the attractive forces. Another effective strategy, often used concurrently, is to control the magnetic particle concentration [11]. By diluting the magnetic phase, the average interparticle distance increases, thereby weakening the strength of the dipole-dipole interactions [12].

A practical method for achieving this dilution in high-solid-content coatings is the incorporation of nonmagnetic spacer particles, such as α-Fe_2O_3 (hematite) or TiO_2, which physically separate the magnetic particles. This approach has been shown to be effective; for instance, Nagai and Inoue demonstrated that diluting the magnetic layer with nonmagnetic particles could successfully reduce media noise while compensating for the lower output [13,14].

To optimize this process, it is crucial to quantify the dispersion quality and the strength of interparticle interactions in the wet ink state, before the coating is finalized. Several characterization techniques are exceptionally well-suited for this purpose.

First, the bulk rheological properties of the suspension, particularly the yield stress (τ_y), are highly sensitive to the formation of a percolated particle network. A higher yield stress directly corresponds to a stronger, more aggregated microstructure [11].

Second, transverse susceptibility (χ_t), which measures susceptibility perpendicular to an applied DC aligning field, is a well-established method for probing the internal magnetic fields (both interaction and anisotropy fields) in magnetic dispersions and solid tapes [6,10,15-18].

Finally, frequency-dependent (AC) complex susceptibility measurements can probe the rotational dynamics of the particles [19]. This method can effectively distinguish between the relaxation behavior of single, isolated particles and that of larger agglomerates, thus providing critical insights into the microstructural state [20,21]. Potanin et al. have also developed theoretical models demonstrating that susceptibility measurements can characterize the degree of particle dispersion quantitatively [12].

In this study, we systematically investigate the impact of replacing magnetic Co-γ-Fe_2O_3 particles with nonmagnetic α-Fe_2O_3 particles on the properties of the wet magnetic ink. The primary objective is to characterize the dispersion quality and quantitatively understand how this dilution controls the interparticle interaction strength. We employ a correlated approach using these three independent measurement techniques: rheological measurements (yield stress, τ_y), transverse susceptibility (χ_t), and complex magnetic susceptibility. By analyzing the corresponding changes in yield stress, the transverse susceptibility peak (H_{peak}), and the magnetic relaxation time (τ), we provide consistent experimental evidence that dilution with nonmagnetic particles effectively reduces interparticle magnetic interactions, breaks down aggregates, and significantly improves the overall dispersion quality.

II. Materials and Methods

Magnetic inks containing a mixture of nonmagnetic (0-50 wt%) and magnetic particles were prepared. The ink consisted of 37 wt% solids (nonmagnetic particles, magnetic particles and polymer) dispersed in cyclohexanone by ball milling for 48 hours. The amount of polymer was 8 wt% of the particle weight. The magnetic particles used were acicular Co-γ-Fe_2O_3 particles (coercivity

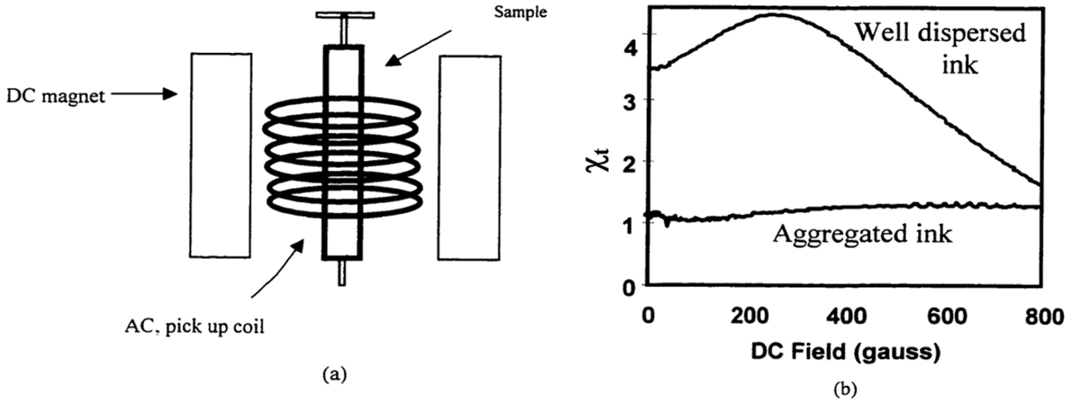

Fig. 1. Schematic of transverse susceptometer (a) and typical example of transverse susceptibility as function of DC field (b).

683 Oe, saturation magnetization 84.8 emu/g, average length 0.3 μm, aspect ratio 6, and BET SSA 30.2 m^2/g). Polyvinylchloride copolymer (MR110, $\overline{Mn}$ = 12,000 g/mol, Nippon Zeon) containing 0.7 wt% SO_4 and 0.5 wt% OH functional groups were used as a wetting resin.

Acicular α-Fe_2O_3 particles were used as a nonmagnetic material. The α-Fe_2O_3 particles were made by heating the Co-γ-Fe_2O_3 magnetic particles at 700°C for 4 hours. TEM measurements showed that these particles were almost the same size and shape as the original particles. X-ray diffraction verified that the alpha phase was formed. The susceptibility and saturation magnetization were about 5% of the original magnetic powder which allowed us to consider the particles nonmagnetic for the purposes of this study.

Transverse susceptibility, χ_t, of the wet inks was measured on an apparatus shown in the reference [6]. It consists of sample holders, pick-up coil, ac coil, and DC magnets as shown in Fig. 1(a). An ink sample is placed in the pick-up coil and initially exposed to a small AC field to move the particles along the AC field. Then, a transverse DC field sweep (0~1,950 Gauss) is applied and the sinusoidal signal generated by the movement of magnetization in ink is reported at each DC field [22].

In Fig. 1(b), a typical graph of dc sweep results is presented. The overall shape of this figure can be explained as a combination of an increasing magnetization (particle orientation) and the pinning of the magnetization with the dc field. The signal is maximum when the long axis of particle aligns with the DC field. This movement is the origin of the increase of the transverse susceptibility at low field. The pinning of the magnetization with the dc field is the origin of the decrease of the χ_t at the high field.

An AC magnetic field is applied to wet magnetic ink while a small AC magnetic field probes susceptibility in a perpendicular direction. χ_t goes through a maximum as a function of the applied DC field. The position of the maximum corresponds to the magnetic field necessary for orienting the magnetic particles. The amplitude and the position of the maximum depend on the magnetic interactions and can be used for characterizing the magnetic inks.

The rheological measurements of magnetic inks were performed using a controlled stress Haake RS-100 rotational rheometer equipped with cone and plate geometry. Cone diameter was 35 mm and cone angle 4°. All measurements were performed in steady shear mode at a temperature of 20°C.

III. Results and Discussion

The dependence of the transverse susceptibility on the applied DC magnetic field is shown in Fig. 2. In this figure, transverse susceptibility is scaled with respect to the volume fraction of magnetic particles because nonmagnetic particle does not contribute to the magnetization. A mean field theory has been developed to explain its behavior. The overall shape of this figure can be explained as a combination of an increasing magnetization (particle orientation) with DC field, which is the origin of the increase of the transverse susceptibility at low field and an increase of the pinning of the magnetization with the DC field, which is the origin of the decrease of the transverse susceptibility at high field. As shown in Fig. 2, the transverse susceptibility at zero DC field depends on the diluted concentration with nonmagnetic particles. The particles are allowed to move even if without the magnetic field and they tend to form agglomerate with different microstructure which depends on magnetic interaction. The decrease of transverse susceptibility with increase of magnetic particle portion at low DC field

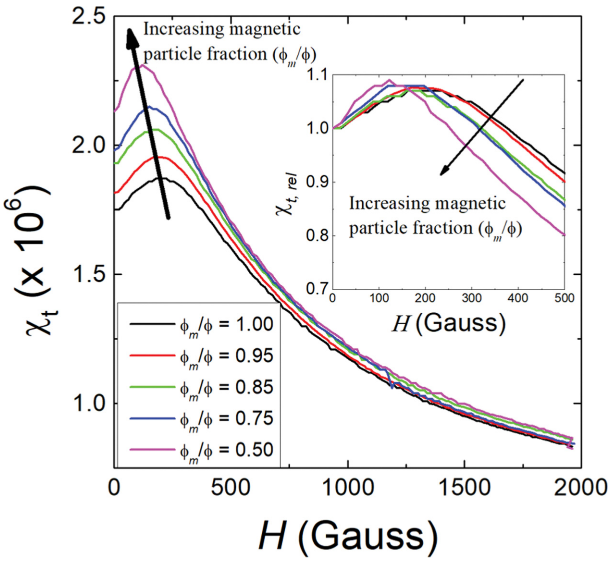

Fig. 2. (Color online) Transverse susceptibility (χ_t) vs. magnetic field strength (H). Inset shows the relative transverse susceptibility ($\chi_{t,rel}$), normalized by the zero-field susceptibility, vs. H.

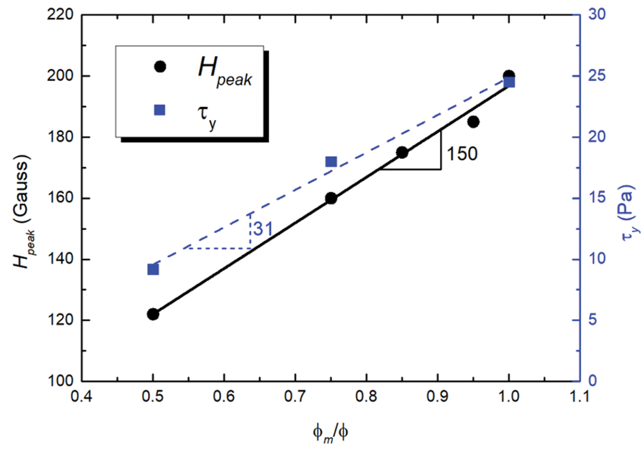

Fig. 3. (Color online) Magnetic field strengths (H_{peak}) for maximum transverse susceptibility and yield stress (τ_y) vs. normalized volume fraction of magnetic particles (ϕ_m/ϕ).

occurs because the magnetic particle interaction field direction is opposite to imposed DC field direction so that the particle orientation is inhibited. At high DC field, the imposed field overcomes the inter-particle magnetic interaction field so that the particles align to field direction.

The inset of Fig. 2 shows the relative transverse susceptibility curve for different dilution concentration of nonmagnetic particles, which is characteristics of magnetic interaction. The maximum susceptibility shifts to lower fields, and its relative amplitude increases with the concentration of the nonmagnetic particles. Such behavior indicates easier orientation of the magnetic particles to the DC field direction. The application of DC field causes the magnetic moments of particles to move away from the easy axis in the direction of the DC field. There is no physical movement for the particles in solid dispersion. But in liquid dispersion, particle orientation is achieved by applying a DC field to the magnetic ink. The particles are free to rotate; the particles orient with their easy axes in the direction of DC field. To consider the effect of inter-particle magnetic interaction on magnetic field, the effective magnetic field $H_{eff}^{DC} = H^{DC} + \beta M \langle \cos\theta_m^{eq} \rangle$, where β is a phenomenological coefficient and θ_m^{eq} is the angle of a local magnetic moment relative to a specific axis at the equilibrium condition. The second term here represents the interaction field. The interaction is positive ($\beta > 0$) if nearest neighbor interactions are such as make parallel alignment of neighboring magnetic moments. The interaction is negative ($\beta < 0$) if particles demagnetize each other. Theoretical simulations based on the mean-field model reported by Potanin et al. [12] demonstrate that the shift of the susceptibility peak to lower fields corresponds to a decrease in the absolute value of the interaction parameter β. This decrease in β signifies weakened demagnetizing interactions among the particles. This theoretical prediction provides a qualitative agreement with our experimental data, where the dilution with nonmagnetic particles effectively reduces the interaction field strength.

Fig. 3 shows that DC field of maximum susceptibility is proportional to the volume fraction of the magnetic particle in magnetic inks and that proportionality is related to the yield stress of magnetic ink, indicating the inter-particle interaction. Diluting magnetic ink with nonmagnetic particles improves the dispersion quality of magnetic ink, as evidenced by a decrease of yield stress and peak of transverse susceptibility. Without shear force, the magnetic particles may form some microstructure in magnetic ink. The higher magnetic field strength required to destroy higher microstructure. To characterize the microstructure, the various forms of microstructures were suggested such as single particle, primary aggregation, which can only be reduced in size by breaking, and network structure formed by magnetically interacting single particles and primary aggregates, which is easily broken and rearranged by a relatively small force. As nonmagnetic particles are increased the primary aggregates become better separated due to insertion of nonmagnetic particles in network structure.

Fig. 4 shows the frequency dependence of the magnetic susceptibility measured at zero DC field on the

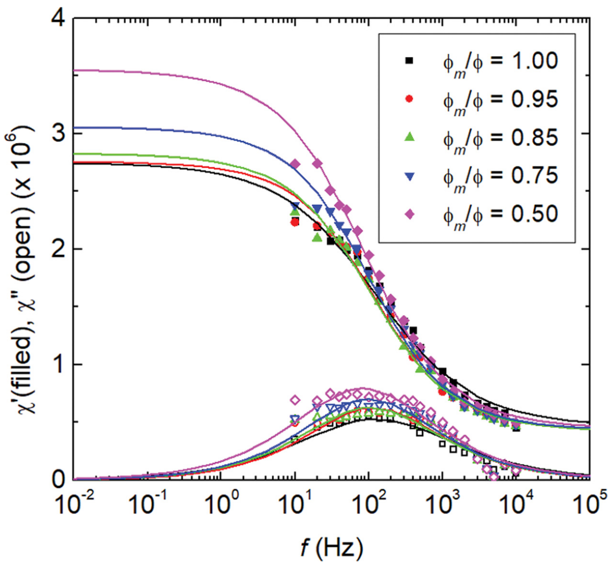

Fig. 4. (Color online) Real (χ') and imaginary (χ'') parts of the complex susceptibility as a function of frequency for various volume fraction of magnetic particles (ϕ_m/ϕ).

concentration of magnetic particles in the inks. The susceptibility measured at high frequency is due only to the displacement of the particle magnetic moment from the easy axis. At low frequencies susceptibility reflects not only the movement of particle magnetic moment but also the physical movement of the particles and in this way probes the small-scale structure of the ink in the immediate vicinity of the magnetic particles. The magnetic analogue of the Cole-Cole plot which is described by the equation

$$\chi(\omega) = \chi_\infty + \frac{\chi_0 - \chi_\infty}{1 + (i\omega\tau)^\alpha} \quad (1)$$

where χ_∞, χ_0 represent the high frequency and zero frequency susceptibilities and α is called the Cole-Cole parameter and τ is the magnetic relaxation time. The low frequency susceptibility and the relaxation time of the susceptibility increase with the nonmagnetic particle portion, suggesting improved dispersion quality. The increase of τ is due to the increased number of single particles in the suspension. The observed increase in both low-frequency susceptibility (χ'_0) and relaxation time (τ) with dilution might initially appear counterintuitive to the weakening of magnetic interactions. However, this behavior is consistent with the structural transition from aggregates to single particles. In the concentrated regime, strong dipolar interactions lock particles into clusters where magnetic flux closure reduces the net susceptibility, and the rotational freedom is constrained, leading to suppressed relaxation dynamics. As nonmagnetic dilution

Table I. The parameters in Cole-Cole plot equation from fitting Fig. 4 data.

ϕ_m/ϕ	**1.00**	**0.95**	**0.85**	**0.75**	**0.50**
χ_0 (× 10^7)	27.5	27.6	28.3	30.6	35.6
χ_∞ (× 10^7)	4.62	4.28	4.20	4.31	4.44
χ_0/χ_∞	5.95	6.45	6.74	7.10	8.02
τ (× 10^3)	8.00	8.60	9.50	9.80	12.00
α	0.35	0.37	0.39	0.38	0.40

weakens these interactions, the aggregates break down into individual particles. These single particles are free to undergo Brownian rotation, which is characterized by a longer relaxation time compared to the constrained dynamics within dense aggregates. Consequently, the increase in τ and χ'_0 serves as a direct indicator of the enhanced mobility and the growing population of single particles, effectively confirming the reduction in interparticle magnetic interactions.

The magnetic susceptibility of the single particles is higher than the susceptibility of the aggregates since large aggregates have small net magnetic moments. As discussed by Potanin et al. [12] and Peikov et al. [23], the susceptibility spectrum shifts to longer relaxation times with the increase of resin concentration due to the steric repulsion and weakening of particle interaction [1,24].

Table I summarizes the Cole-Cole fit parameters extracted from the complex susceptibility spectra in Fig. 4 for the magnetic inks with different normalized magnetic-particle loadings (ϕ/ϕ_m). For each ϕ/ϕ_m = 1.00, 0.95, 0.85, 0.75, and 0.50, the table lists the characteristic relaxation quantities (τ), the characteristic susceptibility amplitude at high and zero frequencies (χ_∞, χ_0), and the broadening parameter (α) from the Cole-Cole model fit. As the magnetic fraction is diluted (lower each ϕ/ϕ_m), χ_0/χ_∞ increases (from 5.95 to 8.02 in the reported units), τ grows (from 8.00 to 12.00 in the reported units), and α shows a slight rise (from 0.35 to 0.40), which is consistent with weakened interparticle interactions upon nonmagnetic dilution.

IV. Conclusion

We have investigated the impact of replacing magnetic Co-γ-Fe_2O_3 particles with nonmagnetic α-Fe_2O_3 particles on the magnetic interactions and microstructure in wet magnetic inks. The objective was to characterize the dispersion quality and understand how dilution controls interaction strength. Our findings from three independent measurement techniques provide a consistent conclusion.

First, transverse susceptibility measurements showed that as the concentration of nonmagnetic particles increased, the magnetic field at peak susceptibility (H_{peak}) shifted to lower applied DC fields. This shift is attributed to a reduction in the negative (demagnetizing) interparticle interaction field, which allows the magnetic particles to be oriented more easily.

Second, rheological measurements demonstrated a proportional decrease in yield stress (τ_y) as the magnetic particle volume fraction (ϕ_m/ϕ) was reduced. This indicates that diluting the system with nonmagnetic particles effectively weakens the networked microstructure formed by magnetic interactions.

Finally, complex susceptibility measurements at zero DC field revealed that both the low-frequency susceptibility (χ'_0) and the magnetic relaxation time (τ) increased with the addition of nonmagnetic particles. This behavior is characteristic of an improved dispersion, specifically reflecting an increase in the number of single particles, which have a higher susceptibility than aggregates.

In summary, all experimental results confirm that diluting magnetic ink with nonmagnetic particles effectively reduces interparticle magnetic interactions. This leads to significantly improved dispersion quality by separating primary aggregates, which is expected to reduce recording noise and improve the overall performance of the magnetic ink.

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Conflict of Interest

The author declares that they have no conflict of interest.

References

[1] D. Farrell, Y. Cheng, R. W. McCallum, M. Sachan, and S. A. Majetich, J. Phys. Chem. B **109**, 13409 (2005).

[2] C. D. Mee and E. D. Daniel, Magnetic Recording Technology, Eds. McGraw-Hill, New York (1996) pp. 6~45.

[3] F. Jorgensen, The Complete Handbook of Magnetic Recording, Tab Books Inc., Blue Ridge Summit (1988) pp. 13 ~25.

[4] R. L. White, J. Magn. Magn. Mater. **209**, 1 (2000).

[5] S. N. Piramanayagam, J. Appl. Phys. **102**, 011301 (2007).

[6] A. A. Potanin, S. M. Shrauti, D. W. Arnold, and A. M. Lane, J. Appl. Phys. **81**, 3803 (1997).

[7] R. E. Rosensweig, Ferrohydrodynamics, Cambridge University Press (1985) pp. 33~72.

[8] D. H. Napper, Polymeric Stabilization of Colloidal Dispersions, Academic Press, London (1983) pp. 18~30.

[9] A. Brunsch, W. Steiner, and G. Trippel, Method and apparatus for characterizing magnetic coating compositions as well as improving magnetic particle dispersion, U.S. Patent No. 4,785,239 (1988).

[10] P. M. Sollis, P. R. Bissell, P. I. Mayo, R. W. Chantrell, R. G. Gilson, and K. O'Grady, J. Magn. Magn. Mater. **120**, 94 (1993).

[11] A. A. Potanin, S. M. Shrauti, D. W. Arnold, and A. M. Lane, Rheol. Acta **37**, 89 (1998).

[12] A. A. Potanin, S. M. Srauti, D. W. Arnold, and A. M. Lane, J. Magn. Magn. Mater. **170**, 298 (1997).

[13] A. A. Potanin, R. J. Hirko, V. T. Peikov, and A. M. Lane, J. Rheol. **42**, 1249 (1998).

[14] N. Nagai and M. Inoue, Abstracts, The 7th Joint MMM-Intermag Conference, San Francisco, California, Jan 6-9 (1998), p 200.

[15] R. Matarranz, M. C. Contreras, G. Pan, B. Presa, J. A. Corrales, and J. F. Calleja, J. Appl. Phys. **99**, 08Q504 (2006).

[16] D. Cimpoesu, A. Stancu, and L. Spinu, Phys. Rev. B **76**, 054409 (2007).

[17] D. Kechrakos and K. N. Trohidou, Phys. Rev. B **74**, 144403 (2006).

[18] S. A. Mathews, C. Musi, and N. Charipar, Sci. Rep. **11**, 3155 (2021).

[19] B. W. M. Kuipers, I. A. Bakelaar, M. Klokkenburg, and B. H. Erné, Rev. Sci. Instrum. **79**, 013901 (2008).

[20] P. C. Fannin, B. K. P. Scaife, and S. W. Charles, J. Magn. Magn. Mater. **72**, 95 (1988).

[21] J. L. Garcia-Palacios, Adv. Chem. Phys. **112**, 1 (2000).

[22] J. P. Embs, A. Leschhorn, and M. Lücke, Phys. Rev. E **73**, 036302 (2006).

[23] V. Peikov and A. Lane, J. Colloid Interface Sci. **206**, 350 (1998).

[24] C. N. Marin, P. C. Fannin, and I. Malaescu, J. Magn. Magn. Mater. **388**, 68 (2015).