SUPPLEMENTAL INFORMATION

Numerical simulations of harmonic magnetizations

We considered spherical single-domain particles whose dynamics in a liquid suspension are dominated by Brownian rotational relaxation. In this case, the behavior of magnetic fluids can be described by the Fokker-Planck equation[1]:

$$2\tau_B \frac{\partial W(\theta,t)}{\partial t} = \frac{1}{\sin\theta} \frac{\partial}{\partial\theta}\left\{\sin\theta\left[\xi(t)\sin\theta \cdot W(\theta,t) + \frac{\partial W(\theta,t)}{\partial\theta}\right]\right\}, \# (S1)$$

where θ is the angle of the magnetic moment m with respect to the excitation field of $H(t) = H_0 \cos\omega t$, $W(\theta,t)$ is the distribution function of m, $\xi(t) = \frac{\mu_0 m H(t)}{k_B T} = \xi_0 \cos\omega t$ is the ratio of excitation field energy to the thermal energy, k_B is the Boltzmann constant, and T is the absolute temperature.

We expand $W(\theta,t)$ into spherical harmonics as

$$W(\theta,t) = \sum_{n=0}^{\infty} a_n(t) P_n(\cos\theta). \#(S2)$$

Where $P_n(\cos\theta)$ are the Legendre polynomials and $a_n(t)$ are time-dependent coefficients for each spherical harmonic. Using some recurrence relations and the orthogonal property of Legendre polynomials, we obtain the following infinite set of differential-difference equations[1].

$$\frac{2\tau_B}{n(n+1)}\dot{a}_n + a_n = \xi(t)\left(\frac{a_{n-1}}{2n-1} - \frac{a_{n+1}}{2n+3}\right). \#(S3)$$

Then, we make the substitution

$$f_n(t) = \int_0^{\pi} P_n(\cos\theta) W(\theta,t) \sin\theta d\theta. \#(S4)$$

Using the orthogonal property of Legendre polynomials, we obtain the following relation between $f_n(t)$ and $a_n(t)$.

$$f_n(t) = \frac{2a_n(t)}{2n+1} \#(S5)$$

Substituting eqn (S5) into eqn (S3) yields the infinite set of differential-difference equations for $f_n(t)$:

$$\tau_B \dot{f}_n(t) + \frac{n(n+1)}{2} f_n(t) = \xi(t) \frac{n(n+1)}{2(2n+1)}\left[f_{n-1}(t) - f_{n+1}(t)\right] \#(S6)$$

As our goal is to calculate the magnetization in the steady state, we expand $f_n(t)$ as a Fourier series

$$f_n(t) = \sum_{k=-\infty}^{\infty} F_k^n(\omega) e^{jk\omega t}, \#(S7)$$

where $F_k^n(\omega)$ are the complex Fourier coefficients. Since all the $f_n(t)$ are real, the complex Fourier coefficients satisfy

$$F_{-k}^{n}(\omega) = \left[F_k^n(\omega)\right]^* \quad \#(S8)$$

where the asterisk denotes the complex conjugate.

Substituting eqn (S7) into eqn (S6), we obtain the following recurrence relations for the complex Fourier coefficients.

$$z_{n,k}(\omega)F_k^n(\omega) - \xi_0\left[F_{k-1}^{n-1}(\omega) + F_{k+1}^{n-1}(\omega) - F_{k-1}^{n+1}(\omega) - F_{k+1}^{n+1}(\omega)\right] = 0 \quad \#(S9)$$

where $z_{n,k}(\omega) = 2(2n+1)\left[1 + j\dfrac{2\omega\tau_B k}{n(n+1)}\right]$.

Using the matrix continued fraction technique[2], we can calculate $F_k^1(\omega)$. The time response of $f_1(t)$ in the steady state can be expressed exclusively in terms of the odd-number components of $F_k^1(\omega)$. Thus, taking eqn (S8) into account, we obtain

$$f_1(t) = \int_0^{\pi} W(\theta,t)\sin\theta\cos\theta d\theta = 2\sum_{k=1}^{\infty} Re\left[F_{2k-1}^{1}(\omega)e^{j(2k-1)\omega t}\right]. \quad \#(S10)$$

The amplitude of (2k-1)-th harmonic of the magnetic nanoparticles, a_{2k-1}, which is given by eqn (9), can be calculated as

$$a_{2k-1} = 2\left|F_{2k-1}^{1}(\omega)\right|. \quad \#(S11)$$

References

1. W. T. Coffey, P. J. Cregg and Y. P. Kalmykov, in Advances in Chemical Physics, edited by I. Prigogine and S. A. Rice, New York: Wiley, 1993, Vol. 83, p. 263.
2. J. L. Déjardin and Yu. P. Kalmykov, Phys. Rev. E, 2000, 61, 1211–1217.