Low oscillatory-field relaxometry for estimating hydrodynamic-size distribution of magnetic nanoparticles dispersed in a liquid medium

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The relaxation behaviors involving both magnetic moment and particle rotations are key phenomena in a dynamically-magnetized magnetic nanoparticle suspension, which are practically observable through its magnetization responses. For a ferrofluid, the varying primary and secondary (hydrodynamic) sizes of the suspended particles are responsible for the broadening relaxation-time distribution which leads to technical disadvantage toward magnetic biosensing (*i.e.*, liquid-phase magnetic immunoassay) in term of accuracy. As an alternative method of preliminary particle-size characterization, we developed a low oscillatory-field relaxometry which is, in principle, an estimation of relaxation-time distribution corresponding to frequency-dependent complex magnetic susceptibility, further addressable to the particle-size distribution. Practically, we implemented a coil-based inductive magnetometry which measures the inductive voltage triggered by a frequency-modulated magnetic field, further converted into a frequency-domain transfer-function to obtain signal components: magnitude and phase. Comparing these parameters of a colloidal magnetic sample to that of the reference (*i.e.*, free sample) in terms of magnitude ratio and phase difference estimates complex magnetic susceptibility of the sample, which is further correlated with the Debye relaxation model to generate a discrete probability density function (PDF) of relaxation-time distribution. The corresponding hydrodynamic-size is then calculated by satisfying the well-known Brownian relaxation-time equation[1)] on each sampling point of the PDF.

To demonstrate the performance of the system, we, here, evaluated two water-based iron oxide nanoparticle suspensions having roughly 0.5 wt. % particle-concentration and different surface-coating; these suspensions are sufficiently-dense to be accurately-characterized by dynamic light scattering (DLS) measurement. A chirp ac current with the logarithmic instantaneous sampling-frequencies ranging from 275 Hz to 325 kHz was fed to an impedingly-controlled induction coil, thus producing a relatively-constant 1 Oe_{rms}-magnetic field at the operating frequency range. Under this synthesized magnetic-field, the frequency-dependent magnetic moments of the 80μl samples were characterized, as shown in Fig. 1a. Since the applied field is considerably-small, thermal energy should be dominant to kinetically-randomize the particle motions, instead of particle rotation. This system, therefore, indirectly measures translational Brownian relaxation, as well as nanoscopic single-particle vibration of the weakly-clustered-particles, in which the resulting magnetization dynamics are coherent with the applied field. Correspondingly, the hydrodynamic-size distribution $\rho(D_h)$ extracted from Fig. 1a indicates that the samples were polydispersive ferrofluids with the overlapped lognormal size-distributions (Fig. 1b). For sample 1 (having 15.85 and 20.01 nm in mean primary-sizes), there are 2 mean secondary-particle-sizes at 34.12 and 66.69 nm, in which the smaller D_h is attributable to single-particle dispersion, whereas the larger one may include the cluster formed. Meanwhile, for sample 2 (having 6.35 nm in mean primary particle-size), we indicated the multiple mean secondary-particle-sizes at 19.11, 35.87, 48.42, and 121.51 nm, suggesting that the suspended particles exist as a particle-cluster. In conclusion, we have shown that a low oscillatory-field relaxometry is capable of analyzing hydrodynamic size distribution of ferrofluid. However, our current system can only characterize magnetic samples which have magnetic moment more than 1×10^{-5} emu.

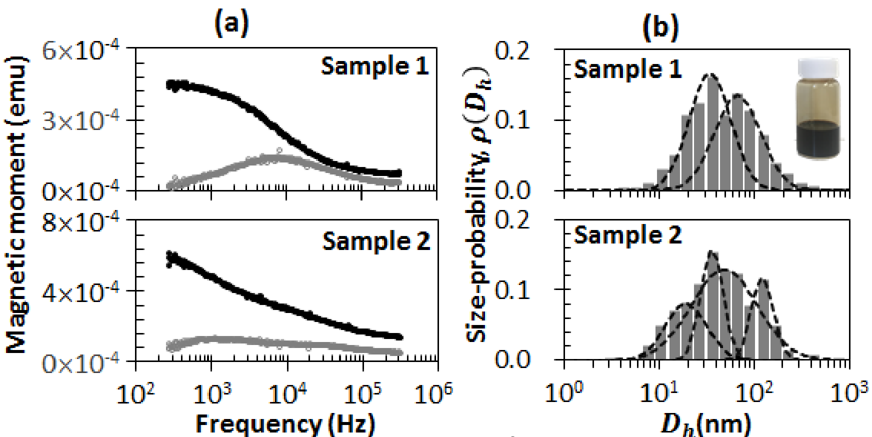

Fig. 1 (a) Frequency-dependent magnetic moment of magnetic suspensions measured at 1 Oe_{rms} and (b) numerical estimation of the corresponding hydrodynamic size distribution of the suspended particles

Reference

1) W. T. Coffey *et al*, Adv. Chem. Phys., **83** (2007) 263.