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THESIS OUTLINE

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【題目】

Characteristic response of electromagnetically stimulated magnetic nanoparticles and its potential application for biosensing
（磁性ナノ粒子の電気的・磁気的励起による応答とそのバイオセンシングに対する応用）

【要約】

As bioanalytical characterization of a colloidal mixture between biomolecular analytes and magnetic nanolabels, wash-free liquid-phase magneto-assay promotes a simplicity in the biosensing mechanism yet offers a comparable accuracy. Aiming for such medical diagnostics system, this doctoral research proposes an effective way to distinguish the analyte-to-label immobilization and any other polydispersive states of magnetic nanolabels (*i.e.*, magnetic particle clustering). This research explicitly highlights the importance of understanding magnetic nanoparticle behavior from the apparent magnetic and electric responses. It suggests a careful-but-rapid measurement of dynamic magnetism in terms of frequency and field-strength dependences, as well as a magnetic field-dependent impedance characteristics. In general, the dissertation is structured into 8 chapters, as follows:

Chapter 1 [General Introduction] describes a technological background of magnetic biosensing, identifies the related problems, and further specifies research objectives with regard to the originality.

Chapter 2 [Theoretical Background and Presumptions] discusses about the concept of a high-speed magnetometry to characterize the magnetic responses of a colloidal superparamagnetic nanosystem. Alternatively, this chapter also puts a stressing point on the origin of magnetic field-induced resistance change in ferrofluids, while introducing a magnetically assisted bipolar impedance spectroscopy as the corresponding electrical characterization method.

Chapter 3 [Magnetic Characterization System and Protocols] introduces the development of coil-based inductive magnetometry to measure frequency and field-strength dependent complex magnetic susceptibility of magnetic nanoparticles with 10^{-5} emu moment sensitivity. This chapter also discusses a rapid magnetic susceptibility measurement involving a discrete Fourier transform of a frequency-modulated magnetic field-pulse to minimize the external noises and harmonics. The corresponding new analytical protocols of the measured susceptibility are formulated to estimate hydrodynamic size, the medium viscosity, as well as static and dynamic magnetization including moment distributions.

Chapter 4 [Magnetic Properties of Ferrofluid at Low Magnetic-field Regime] details the experimental study on relaxation dynamics of polydispersed-particle systems through low oscillatory-field relaxometry.

The size-dependent spectral magnetization responses (*i.e.*, Brownian and Néel relaxations) are found contributing significantly to defining the finite superparamagnetism at low field-regime. The hydrodynamic polydispersity which results in a multimodal relaxation time distribution, can be accurately estimated with regard to the nonlinear relaxation response (*i.e.*, non-Debye model). Moreover, it is found that the interparticle interactions and thermal agitation significantly influence the relaxation dynamics by inducing spectral shift of the effective relaxation frequency.

Chapter 5 [Rotational Brownian Kinetics of Magnetic Nanoparticles] discusses about the significant contribution of magnetodynamic moment non-uniformity in Brownian relaxation dynamics, in addition to the medium viscosity and thermal effects. The magnetodynamic moment which is treated as an effective magnetic moment in a cluster, in principle, is introduced as a unit of an oscillatory magnetic field-induced magnetization-response of a magnetic nanoparticle system forming clusters in liquid medium, thus it is sensitive to dipolar interparticle interaction. Depending of the applied frequency, magnetodynamic moment at low frequency region reflects the magnitude of magnetic moment of the cluster corresponding to the hydrodynamic diameter, whereas in the high frequency region, it represents the magnitude of magnetic moment of individual magnetic nanoparticles, with regards to Néel relaxation and non-rotatability of the particles.

Chapter 6 [Electrical Impedance Characteristics of Ferrofluid] provides an investigative detail regarding the impedance characteristics of a ferrofluid under external magnetic field, including the suitable electrode design and possible measurement artifacts (e.g., volumetric change, thermal disturbances, electrode polarization, and inductive noises). The change in electrical resistance found in a relatively dense ferrofluid sample, seems to have a field-strength dependence upon the applied dc and ac magnetic fields. The origin of this magnetic field-induced resistance change is related to the temporal chain-like particle aggregation under dc field and the particle rotation under ac field. It was also found that electromagnetic phase-difference is an important factor to observe the resistance change under ac magnetic field. Nevertheless, both ac and dc magnetic fields induce a trivial change in electrical properties, which is not practically suitable for magnetic biosensing

Chapter 7 [Wash-free Liquid-phase Magneto-assay] discusses a laboratorial trial on detecting three types of analytes: ions, electrically charged amino acid, and polymeric macromolecule, by employing the non-specific electrostatic analyte-immobilization on to magnetic labels. The rapid measurement by using the low oscillatory-field relaxometry and multi-frequency-mode dynamic magnetometry seem successfully capable of distinguishing the change in of frequency and field-strength dependent magnetic susceptibility of the ferrofluid sample as it mixes with the analyte solution. The corresponding cross-analysis of hydrodynamic size and magnetodynamic moment distributions further shows its effectiveness in evaluating the polydispersive state of magnetic labels. This method, therefore, is suitable for developing wash-free liquid-phase magneto-assay

Chapter 8 [General Conclusions] summaries the principal research achievements and specifies the remaining challenges and recommendations.