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**THESES OF THE DOCTORAL
DISSERTATION**

**Examination of electric and magnetic
properties of intelligent materials**

(Intelligens anyagok nemlineáris mágneses és elektromos
tulajdonságainak vizsgálata)

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1 Abstract

Intelligent materials are functional systems that, under external excitation (electric, magnetic, mechanical, or thermal stimuli), rapidly and reversibly change their material properties. The present dissertation focuses on an important subset of intelligent materials, magnetic fluids, and investigates their linear and nonlinear dynamic magnetic behavior in alternating (AC), and combined alternating and direct (AC+DC) magnetic fields. The investigations center on the frequency-dependent complex susceptibility and its higher-order (harmonic) components, which provide direct information on relaxation processes, interactions, and saturation nonlinearity.

During the experimental work, a self-developed mutual-induction (differential-transformer) AC susceptometer was realized. The differential connection of the reference and sample coils, precision current-source excitation, and lock-in detection make it possible to reproducibly determine the spectrum of the complex susceptibility $\chi^*(\omega)$. The measurement system was validated using commercial magnetic fluids: Ferrotec EMG700 and EMG900 ferrofluids, and the Parker MRF-140CG magnetorheological fluid. For ferrofluids, the spectra show characteristic relaxation behavior; for the quantitative characterization of the data, Cole–Cole type fitting was applied, and based on temperature-dependent measurements, activation energy estimates were performed. In the case of the magnetorheological fluid, in addition to the low-frequency, nearly quasi-static range, high-frequency losses and distortions of the measurement system constitute key aspects of interpretation.

In the theoretical part, based on the Mean Spherical Approximation (MSA) description of the dipolar hard-sphere (DHS) model, the

implicit magnetization equation of Szalai and Dietrich was expanded in powers of the field strength up to seventh order, and closed-form expressions were provided for the nonlinear coefficients. Using the series expansion and a Debye-type relaxation approximation, analytical formulas were derived for the harmonic components of the dynamic susceptibility in a purely alternating field, and the theory was then extended to AC+DC excitation. Numerical comparisons show that the seventh-order series expansion provides a reliable approximation in weak and moderate fields relevant for engineering and experimental practice (typically $H_0^* < 1.4$), whereas in strong fields, due to saturation nonlinearity, the harmonics are suppressed after passing through characteristic maxima. DC premagnetization, through symmetry breaking, also generates odd harmonics, and for a large DC ratio it practically “switches off” the nonlinear response; this phenomenon can be directly exploited, among others, in the spatial control of magnetic particle imaging (MPI) and magnetic hyperthermia.

Keywords: intelligent materials; magnetic fluid; ferrofluid; magnetorheological fluid; complex susceptibility; nonlinear response; harmonics; MSA; lock-in detection; AC+DC excitation.

2 Introduction, objectives

The development and investigation of intelligent materials is a dynamically developing area of materials science. These materials and systems, under external excitation (for example in an electric or magnetic field, under mechanical loading, or due to a change in temperature), are able to change their structure and thereby their macroscopic properties. Due to controllability, fast response time, and integrability, intelligent materials appear in numerous engineering applications as sensors, actuators, and adaptive structural elements.

The topic of the dissertation is related to magnetic fluids among intelligent materials. Ferrofluids and magnetorheological (MR) fluids are dispersed systems in which magnetic particles are distributed in a carrier medium, and under the action of an external magnetic field the effective properties of the system can change significantly. In the case of magnetic fluids, the magnetic field not only creates an equilibrium magnetization, but due to alternating excitation it also excites dynamic phenomena: the orientation of the particles and the relaxation of magnetic moments are frequency-dependent, and the response involves phase lag and dissipation. This behavior is described by the complex magnetic susceptibility $\chi(\omega)$.

In a significant part of practical applications, however, the linear approximation valid for small fields is not sufficient. Under moderate and strong excitation, saturation effects, dipole–dipole interactions, and sample-structural effects produce a nonlinear response, one of the most important consequences of which is the appearance of harmonics. The nonlinear dynamic response plays a key role in several modern technologies as well: in magnetic particle imaging (MPI), the signal is based on nonlinear magnetization, while in magnetic hyperthermia,

active control of losses and heating efficiency is often realized by DC pre-magnetization.

For these reasons, the investigation of the dynamic susceptibility of magnetic fluids requires an experimental and theoretical toolkit that reproducibly measures the spectrum of the complex susceptibility, is capable of handling the harmonic components of the nonlinear response, can be related to physical parameters (such as density, dipole moment, temperature), while from an engineering point of view also provides usable, simple models.

The main objectives of the dissertation can be summarized as follows:

1. Construction of a mutual-induction-based AC susceptometer with lock-in detection and development of a measurement protocol for determining the complex susceptibility of magnetic fluids.
2. Validation of the measurement system and interpretation of the spectra of magnetic fluids (ferrofluids and an MR fluid), with special emphasis on relaxation behavior, non-Debye character, and measurement limitations.
3. Development of an MSA-based (Mean Spherical Approximation) theoretical description in a purely alternating magnetic field: series expansion of the magnetization equation in the field strength, followed by the analytical determination of the harmonics of the nonlinear dynamic susceptibility within a Debye-type relaxation approximation.
4. Extension of the theoretical model to AC+DC excitation, investigation of symmetry breaking and the harmonic structure induced by the DC field, and quantitative characterization of the validity range of the series expansion.

3 Thesis statements

1. **Development of the measurement method** I developed and validated a self-built mutual-induction (differential-transformer) AC susceptometer and measurement protocol, in which the differential coil arrangement, lock-in detection, and precision current-source excitation enable reproducible determination of the complex susceptibility spectrum; during validation I also established evaluation procedures applicable in practice (e.g., Cole–Cole fitting, activation energy estimation from temperature dependence) and documented interpretation aspects arising from measurement limitations.

Related publications: [K1], [C1]

- (a) *Integration of lock-in measurement:* by differential coupling of the secondary coils, I applied the phase- and amplitude-sensitive detection of the lock-in amplifier directly to the signal sensitive to the sample effect, which yields stable, reproducible complex (real+imaginary) measurement results.
- (b) *Measurement ranges and sample-specific limitations:* I adapted the measurement settings to the physical characteristics of magnetic fluids (for ferrofluids typically a low-frequency range and small field strength to avoid structure formation).
- (c) *Evaluation protocol:* I documented the steps for determining the complex susceptibility spectrum (amplitude and phase information relative to the excitation) and provided practical checks by which measurement distortions (e.g., system effects) can be separated.

- (d) *Validation on real samples:* I validated the measurement system using commercial magnetic fluids (Ferrotec EMG700 and EMG900 ferrofluids, and the Parker MRF-140CG MR fluid), and presented the main characteristic features of spectrum interpretation.
- (e) *Extraction of relaxation parameters:* for quantitative characterization of ferrofluid spectra, I applied Cole–Cole fitting, and furthermore estimated activation energy from temperature-dependent measurements, linking it to relaxation mechanisms and possible structural effects.

2. Theoretical model in an alternating field I expanded the MSA-based (Szalai–Dietrich) magnetization equation of the dipolar hard-sphere model in the field strength up to seventh order and provided closed-form expressions for the coefficients; then, by incorporating Debye-type relaxation, I derived analytical formulas for the harmonic components of the nonlinear dynamic susceptibility, and numerically identified the validity range of the approximation (it is well usable up to the fourth harmonic in weak fields, while convergence deteriorates in stronger fields).

Related publications: [K2], [C2]

- (a) *Relation to physical parameters:* I documented the MSA-based derivation of the dipolar hard-sphere model and showed that the coefficients of the series expansion are directly related to physical parameters (e.g., density, dipole moment, temperature).
- (b) *Seventh-order series expansion:* I expanded the implicit equation into a series up to seventh order; due to symmetry, spontaneous magnetization and even-power terms vanish,

so the relevant nonlinear coefficients m_1, m_3, m_5, m_7 are obtained.

- (c) *Static–dynamic relation and harmonics*: I provided the formal relation by which the Fourier components of the dynamic response (0th, 2nd, 4th, and 6th harmonics) can be obtained analytically from the field-dependent (static) susceptibility, and I presented the field-strength scaling of harmonic amplitudes.
- (d) *Incorporation of Debye-type relaxation*: I extended the static MSA results to the complex, frequency-dependent case using Debye-type relaxation such that the static coefficients become complex quantities.
- (e) *Convergence and validity range*: I numerically analyzed the convergence of the series expansion, verified that the approximation is excellent in weak fields, that higher harmonics are more sensitive in moderate fields, and that convergence deteriorates with increasing harmonic order.

3. Series-expanded model in an AC+DC field I generalized the description based on the seventh-order MSA series expansion to AC+DC excitation and provided analytically usable spectrum formulas for harmonics with $n \leq 4$; by numerical comparison I verified that the approximation is reliable up to moderate field strength ($H_0^* < 1,4$, typically 0–25 kA m⁻¹), and I showed that DC bias generates odd harmonics through symmetry breaking and, for large DC ratio, the nonlinear response can be purposefully “disabled”.

Related publication: [C3]

- (a) *Excitation form and symmetry breaking:* in the AC+DC case the magnetic field has the form $H(t) = H_{\text{dc}} + H_0 \cos(\omega t)$. The DC component causes symmetry breaking, which leads to a new harmonic structure.
- (b) *Harmonic complex susceptibilities:* from the series-expanded theory, the complex susceptibilities of the linear and $n = 1 \dots 4$ harmonics ($\hat{\chi}_0, \hat{\chi}_\omega, \hat{\chi}_{2\omega}, \hat{\chi}_{3\omega}, \hat{\chi}_{4\omega}$) can be computed directly using analytical formulas.
- (c) *Accuracy relative to an exact reference:* by quantitative comparison I demonstrated that for a typical bias ratio (e.g., $H_{\text{dc}}^*/H_0^* = 0,2$) at $H_0^* \approx 1$ the spectra of the series expansion and the exact solution are practically identical (with deviations below 1% for the examined components).
- (d) *Validity range:* I specified the range in which the series expansion remains reliable ($0 \leq H_0^* \leq 1,4$), and by converting this to experimental parameters I also connected it to the typical field range of 0–25 kA m⁻¹.
- (e) *Identification of limitations of the series expansion:* I pointed out that at higher field strengths the series expansion may exhibit artifacts (local maxima, oscillations) that do not appear in the exact solution; therefore, in strong fields the approximation cannot be applied without correction.

4. **Exact model in an AC+DC field** I determined the exact time dependence of magnetization by an iterative numerical solution of the implicit MSA equation under excitation $H(t) = H_{\text{DC}} + H_{\text{AC}} \cos(\omega t)$, then treated the resulting time dependence (as a real signal) and transformed it into a spectrum by Fourier analysis, from which I computed the harmonic susceptibilities. I used the exact solution as a reference basis for the quantitative

verification of the accuracy of the series-expanded description and for interpreting the strong-field saturation characteristics.

Related publication: [C3]

- (a) *Generation of the exact time dependence:* I determined the MSA time dependence numerically under AC+DC excitation and presented the time evolution of $H(t)$ and $M(t)$ for different parameters.
- (b) *Spectrum formation and harmonics:* from the magnetization time signal I formed a spectrum by fast Fourier transform; from its amplitudes I calculated the contributions of the fundamental component and harmonics, and from these the harmonic susceptibilities.
- (c) *Use as a reference:* I used the exact spectrum to verify the accuracy of the series-expanded formulas, and thereby quantitatively supported the applicability of the series expansion in weak–moderate fields and the emergence of saturation nonlinearity in stronger fields.

4 List of related publications

Journal articles

[C1] Horváth, B., Decsi, P. and Szalai, I. (2022). Measurement of the response time of magnetorheological fluids and ferrofluids based on the magnetic susceptibility response. *Journal of Intelligent Material Systems and Structures*, 33(7), 918-927. <https://doi.org/10.1177/1045389X211038697>

[C2] Horváth, B., Decsi, P. and Szalai, I. (2022). Nonlinear contributions to the dynamic magnetic susceptibility of magnetic fluids. *Journal of Molecular Liquids*, 359, 119279. <https://doi.org/10.1016/J.MOLLIQ.2022.119279>

[C3] Horváth, B., Guba, S., Decsi, P. and Szalai, I. (2026). Nonlinear dynamic susceptibility of magnetic fluids insymmetry breaking dc magnetic fields. *Journal of Molecular Liquids*, (Under review, manuscript number: MOLLIQ-D-25-05858)

Conference proceedings

[K1] Decsi, P. and Szalai, I. (2020). Alacsony frekvenciás AC szuszceptométer mágneses folyadékok komplex szuszceptibilitásának mérésére. In I. Barabás (Ed.), XXI. Energetika-Elektrotechnika - ENELKO és XXX. Számítástechnika és Oktatás - SzámOkt Multi-konferencia (pp. 29-33). Erdélyi Magyar Műszaki Tudományos Társaság - EMT.

[K2] Horváth, B., Decsi, P. and Szalai, I. (2022). Nonlinear dynamic susceptibility of magnetic fluids in symmetry breaking dc magnetic fields. In: Chemistry, Physics and Biology of Colloids and Interfaces: Book of Abstracts pp. 40-40. , 1 p.