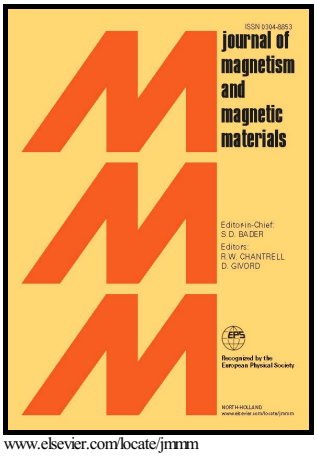

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The influence of interparticle correlations and self-assembly on the dynamic initial magnetic susceptibility spectra of ferrofluids

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Abstract

Using computer simulations and a mean-field theoretical approach, we study how the growth in dipolar interparticle correlations manifests itself in the frequency-dependent initial magnetic susceptibility of a ferrofluid. Our recently developed theory gives the correct single-particle Debye-theory results in the low-concentration, non-interacting regime; and it yields the exact leading-order contributions from interparticle correlations. The susceptibility spectra are analysed in terms of the low-frequency behaviours of the real and imaginary parts, and the position of the peak in the imaginary part. By comparing the theoretical predictions to the results from Brownian dynamics simulations, it is possible to identify the conditions where correlations are important, but where self-assembly has not developed. We also provide a qualitative explanation for the behaviour of spectra beyond the mean-field limit.

Keywords: dynamic initial magnetic susceptibility, dynamic spectra, dipolar interaction, polydispersity, magnetic nanoparticles

1. Introduction

Magnetic AC susceptometry is a widely used technique for analysing and characterising the dynamic magnetic response in ferrofluids [1, 2, 3, 4]. The dynamic response of magnetic-nanoparticle suspensions is fundamentally important for medical applications in general [5], and in particular for magnetic hyperthermia, usually applied alone or in combination with other treatments to eliminate tumours [6]. The efficiency of the latter methods relies on the frequencies and the characteristic relaxation times of the magnetic nanoparticle systems [7, 8, 9]. The influence of magnetic dipolar interactions on the specific absorption rate leads to a decrease or an increase in the hypothermia efficiency depending on the magnetic particle size [10, 11, 12]. This observation has been confirmed in other investigations [13, 14] and it is clear that interparticle correlations should be taken into account when predicting the dynamic susceptibility spectra of magnetic nanoparticles. Until recently, though, a theoretical formalism to allow for correlations was missing. To address this, we have put forward a new theoretical approach [15] based on the analytical solution of the Fokker-Planck equation with an additional term allowing for the interparticle interactions and system polydispersity. We have tested our theory against Brownian dynamics computer simulations of a model monodisperse ferrofluid [16] and against experimental measurements for true polydisperse ferrofluids [17]. In these studies, it was shown that at low temperature and/or high magnetic phase concentration, some complex internal structure developed in the systems, leading to a dramatic increase of the characteristic relaxation times. About 30 years ago, it was suggested that at very low temperatures, the magnetic nanoparticles undergo dynamic arrest and enter a dipolar glass phase [18, 19], but this does not account for typical ferrofluid behaviour.

In the present study we identify how internal structure affects the susceptibility spectrum of a monodisperse system of magnetic dipolar particles by comparing our theory to the results of Brownian dynamics simulations for a broad range of temperatures and concentrations.

2. Problem

Consider the simplest case of a weak probing AC magnetic field $\mathbf{H}(t)$ applied along the symmetry axis (Oz-direction) of a highly elongated cylindrical ferrofluid sample: $\mathbf{H}(t) = (0, 0, h\exp(i\omega t))$; h and ω are the field amplitude and oscillation frequency, respectively. The orientation of each particle's magnetic moment $\mathbf{m}$ is described its polar angle θ with respect to the external field $\mathbf{H}$. A common approach for describing the dynamic magnetic response in ferrofluids is based on the Fokker-Planck-Brown (FPB) equation [20, 21, 22] for the normalised orientational probability density $W(t, \theta_1) \equiv W(1)$ of a randomly chosen magnetic nanoparticle 1.

$$2\tau_1 \frac{\partial W(1)}{\partial t} = \frac{1}{\sin\theta_1}\frac{\partial}{\partial\theta_1}\left\{\sin\theta_1\left[\frac{\partial W(1)}{\partial\theta_1} - \frac{\partial U(1)}{\partial\theta_1}W(1)\right]\right\}, \quad (1)$$

where τ_1 is the characteristic relaxation time of the ferroparticle magnetic moment, which depends on the ferroparticle size, and $U(1)$ is the interaction energy in units of the thermal energy $k_\mathrm{B}T$. Traditionally, only the Zeeman particle-field interaction is considered and so $U(1) = \mu_0(\mathbf{m}_1 \cdot \mathbf{H})/k_\mathrm{B}T$, where μ_0 is the vacuum magnetic permeability. The advantage of this approach is

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