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Preparation and magnetisation of a silica-magnetite inverse ferrofluid

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Abstract

We introduce an 'inverse ferrofluid' comprising sterically stabilized, colloidal silica spheres and oleic acid stabilized magnetite particles. The preparation is described as well as magnetisation measurements which turns out to be a linear function of the silica volume fraction.

Keywords: Inverse ferrofluids; Magnetic holes; Silica; Magnetisation measurements

1. Introduction

'Inverse ferrofluids' are suspensions of nonmagnetic particles in a ferrofluid. When the nonmagnetic particles are much larger than the ferrofluid particles the ferrofluid acts as a continuous, magnetisable medium. In a magnetized ferrofluid the nonmagnetic particles form 'magnetic holes' with a magnetic dipole moment m. For spheres of radius R,

$$\boldsymbol{m} = -\tfrac{4}{3}\pi R^3 \boldsymbol{M}, \tag{1}$$

where $\boldsymbol{M}$ is the magnetization of the ferrofluid. The particles interact via a dipole–dipole potential:

$$U = \frac{\mu_0 m^2}{4\pi r^3}(1 - 3\cos^2\theta). \tag{2}$$

Here r denotes the interparticle distance and θ the angle between the magnetic field and the center to center vector of the dipoles.

Nonmagnetic particles are available with well defined shape and size, and consequently well defined (dipolar) interactions. Skjeltorp [1] introduced the use of polystyrene latex spheres and studied aggregation, crystallization and chain formation in a two dimensionally confined inverse ferrofluid. Popplewell et al. have studied magnetorheological properties using glass beads [2]. Both systems are in the micrometer size range. In the context of a rheological study [3] we are interested in smaller particles which exhibit thermal motion and thus constitute a 'true' colloidal suspension of nonmagnetic particles. We explored the use of small monodisperse silica spheres. An additional advantage of silica is the variety of well-documented chemical surface modifications [4,5] that enable

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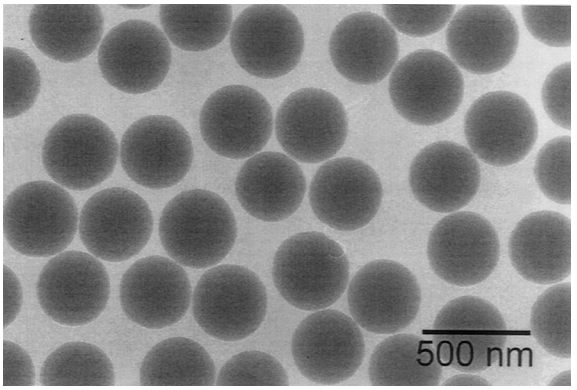

Fig. 1. Electron micrograph of SJL29St. The particles shown here have an average radius of 177 nm, the polydispersity being 4%.

Table 1
Characterization of inverse ferrofluid constituents

	Technique	Parameter	Result
SJL29St	SLS	Radius (nm)	190 ± 15
	DLS	Radius (nm)	205 ± 5
	TEM	Radius (nm)	177
	TEM	Polydispersity	4%
		Density ($gr.ml^{-1}$)	1.804
Ferrofluid	Magnetometry	Saturation magnetization (10^4 A m^{-1})	4.52
	TEM	Radius (nm)	7

dispersibility of silica particles in various organic solvents (e.g. ferrofluids). We found that stearylalcohol-coated silica spheres are stable in ferrofluids based on oleic-acid-coated magnetite particles. In this paper we focus on the preparation method and magnetization measurements.

2. Sample preparation and characterization

2.1. Particle synthesis

Silica particles were prepared by condensation polymerization of tetreaethoxysilane following Stöber [6]. The particle radius was determined using static (SLS) and dynamic light scattering (DLS). The silica particles were coated with stearylalcohol according to van Helden et al. [4]. Particles coded SJL29St from the final, stable suspension in cyclohexane were characterized by electron microscopy (TEM), Fig. 1. Their mass density was measured by determination of the mass fraction silica of the suspension and its density (KEM DA200 densitometer).

2.2. Ferrofluid

The ferrofluid consisted of magnetite particles stabilized by oleic acid in decalin. The ferrofluid was characterized by electron microscopy and magnetization measurements. Characterization results are shown in Table 1. Using Eqs. (1) and (2) and $M_s = 45.2\,\mathrm{kA\,m^{-1}}$ the maximum dipolar interaction energy for two particles at contact turns out to be $1500\,kT$.

2.3. Sample preparation

Silica was dried at 70°C in an oven. Samples were prepared by dispersing a known weight of dry silica in ferrofluid using 30 min of ultrasonication. Stability was checked by visual observation. We found that the solvents decalin, cyclohexane and *n*-hexadecane all gave stable inverse ferrofluids, without any indication of depletion-induced attraction due to the size difference between stearylsilica and ferrofluid particles.

3. Magnetization measurements

Magnetization measurements on (inverse) ferrofluids were carried out as a function of the volume fraction silica, using a MicroMag 2900 alternating gradient magnetometer (Princeton Measurements Corporation) [7] and a glass capillary as a sample holder. Results were checked for independence on the aspect ratio of the fluid cylinder. Magnetization curves are shown in Figs. 2 and 3. The data from Fig. 2 collapse upon a master curve when scaled by the saturation magnetization M_s. This result can be explained by an internal structure of particle chains, of the type also observed by Skjeltorp [1]. Chain formation is due to the dipolar interaction between the constituting particles. These chains can

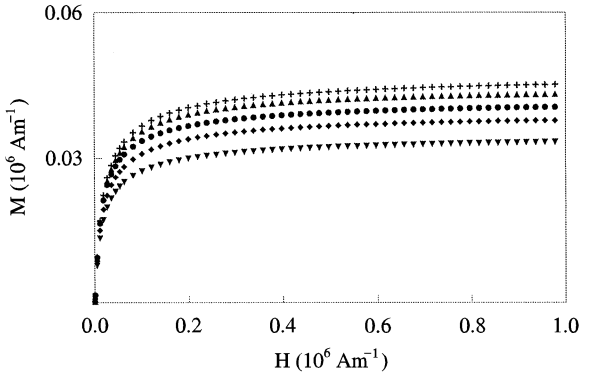

Fig. 2. Magnetization curves of inverse ferrofluid as a function of the volume fraction silica particles (Φ_v: + = 0.021, ▲ = 0.044, ● = 0.102, ◆ = 0.163, ▼ = 0.261)

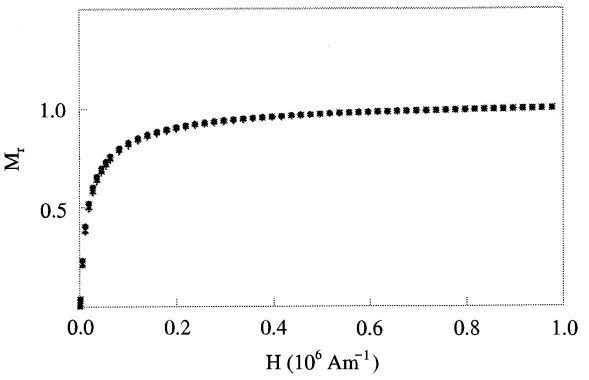

Fig. 3. Reduced magnetization M_r obtained by scaling M in Fig. 2 by M_s.

to a first approximation be described as non-magnetizable cylinders, parallel to the external field H. As the tangential component of H is continuous across interfaces, H is the same everywhere in the sample. Consequently, the dependence of M on the volume fraction silica Φ_v should be

$$M(H,\Phi_v) = M(H)(1 - \Phi_v). \quad (3)$$

As M_s is proportional to $(1 - \Phi_V)$ scaling by M_s indeed should result in a master curve.

4. Conclusions

We have developed a method to prepare stable inverse ferrofluids based on monodisperse sterically stabilized silica particles and oleic-acid-coated magnetite particles. The magnetization of a decalin based inverse ferrofluid as a function of the volume fraction silica follows a single master curve through scaling with the saturation magnetization, implying that $M(H,\Phi_v) = M(H)(1 - \Phi_v)$. This result can be understood in terms of an internal structure of particle chains, which can be modeled as cylinders oriented parallel to the magnetic field.

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