INVESTIGATIONS OF A Fe_3O_4-FERROFLUID AT DIFFERENT TEMPERATURES BY MEANS OF MAGNETIC MEASUREMENTS

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We present results on the investigation of a Fe_3O_4 ferrofluid based on heavy water, using magnetic measurements. Magnetization curves were generated with an original setup at the Department of Magnetism, Moscow State University. As temperature decreases, the value of the saturation magnetization of the ferrofluid increases. The analysis is carried out using the magnetization curves at the temperatures of 80K and 300K. For the two values of the temperature, we reconstruct the lognormal distribution function of particles diameters. We found a higher value of the mean magnetic diameter and a significantly reduced spread in the values of the diameters at the lower temperature.

Se prezintă investigații asupra unui ferofluid Fe_3O_4 în apă grea, folosind metode magnetice de măsurare. Curbele de magnetizare au fost obținute la temperaturile de 80K şi 300K cu un aranjament experimental original, dezvoltat la Departamentul de Magnetism al Universității de Stat din Moscova. Cu scăderea temperaturii, valoarea magnetizației de saturație a ferofluidului studiat creşte. Pentru cele două valori ale temperaturilor s-au reconstituit funcțiile de distribuție lognormală ale diametrelor particulelor. S-a observat că valoarea medie a diametrului particulelor este mai mare şi împrăştierea diametrelor particulelor este mai redusă pentru temperatura coborâtă.

Keywords: magnetization curves, Langevin model, Fe_3O_4 ferrofluid

1. Introduction

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A ferrofluid, also known as a magnetic fluid, is a colloidal suspension of magnetic nanoparticles dispersed in a fluid media, coated with nonmagnetic layer (surfactant or stabilizer) to prevent particle-to-particle agglomeration [1,2]. The most used magnetic material is the magnetite (Fe_3O_4) or the maghemite (γ-Fe_2O_3), obtained by chemical synthesis or by physical methods, while the carrier liquid is typically an oil or water [3,4]. Various methods of coating particles to assure the long-term stability with regard to the temperature and the presence of magnetic interaction have been described in [3]. Since their discovery in 1960s, due to the remarkable variety of magnetic behaviors the ferrofluids continue to present high interest in various applications from chemistry and biology, from data storage and recordings to medical imaging and medical treatment [5,6].

The magnetic properties of ferrofluid systems are interesting not only in the development of a large spectrum of applications but also from a fundamental point of view. The investigation of a system of ultrafine particles and clusters allow the study of the influence of the finite size of the system on the material properties. Nanoscale clusters make a bridge between the bulk system and the atom and their study may show how the bulk properties evolve from the atomic properties when increasing the number of atomic clusters.

In this paper we report results on the investigation of a Fe_3O_4 ferrofluid based on heavy water, using magnetization measurements for the temperatures of 80K and 300K. Magnetic microstructure studies of similar type of ferrofluid by means of nuclear methods were previously carried out and were presented in [7-15].

2. Experimental data

Details on the technique used to the synthesis of the Fe_3O_4 ferrofluid nanoparticles are presented in [7]. In this study we focus on the magnetic properties of this ferrofluid at two different temperatures: 80K and 300K.

Magnetization curves were measured with LakeShore N7400 Vibrating Sample magnetometer at the Department of Magnetism, Lomonosov Moscow State University. The measurements were made in magnetic fields till 16 kOe at the temperature range from 80 up to 300K.

In Fig.1 two magnetization curves for T=80K (circles) and T=300K (squares) are presented. It is observed that the saturation magnetization is lower for the higher temperature. In temperature range from 270 up to 80K the magnetic moment of the sample changes negligibly.

Details on the region of small magnetic fields are obtained by magnification of the linear regions in the interval of the magnetic field intensity between -1500Oe and 1500Oe as shown in Figure 2 where the experimental magnetization curves reveal the hysteresis cycles.

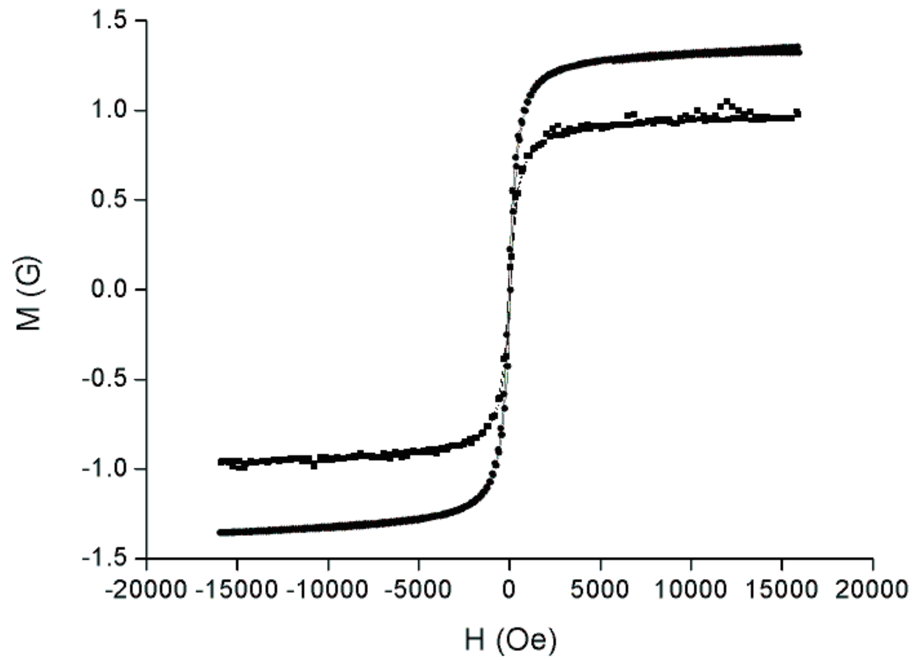

Fig.1 The magnetization as function of the field at the temperatures 80K (circles) and 300K (squares)

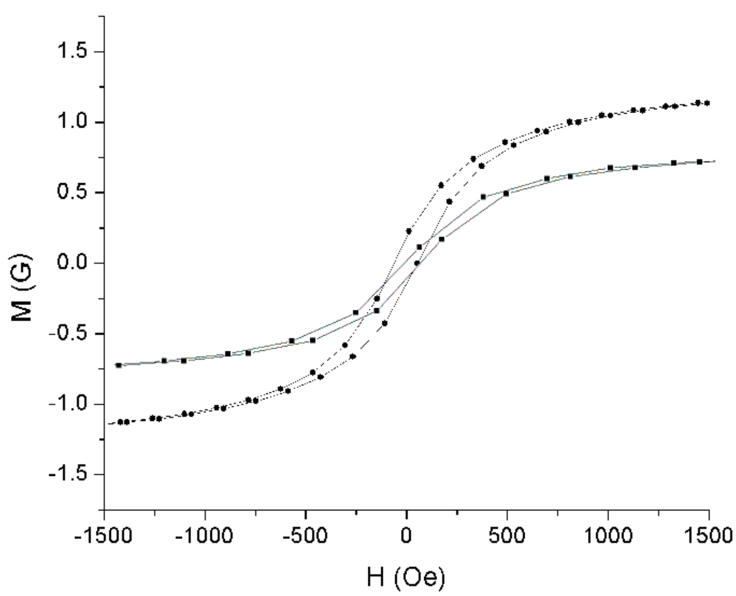

Fig.2 Magnification of the linear region of the magnetization curves: T=80K (circles) and T=300K (squares)

3. Results and discussions

According to the Langevin model of paramagnetism, the ferrofluid is considered as composed by non interacting spheres with a permanent dipolar magnetic moment (*m*) rotating together with the particle when applying an external magnetic field. The macroscopic magnetization is obtained as a result of the combined action of an orientation induced by the external field and a destabilization produced by the Brownian motion. In this ideal model, the magnetization is $M(H) = M_{sat} L(\xi)$ where $L(\xi) = \coth(\xi) - 1/\xi$ is the Langevin function of variable $\xi = \mu_0 mH / k_B T$ with μ_0 vacuum permeability, k_B Boltzmann constant, T temperature and M_{sat} the saturation magnetization of the ferrofluid.

Considering the correction related to the contribution of the particles of different dimensions, the distribution function of the particle size (i.e. diameter *D*) must be introduced in the calculus of the magnetization [16]:

$$M(H) = n \int_0^{\infty} m(D) L(\xi(D)) f(D) dD \tag{1}$$

where *n* is the particle number density and *f(D)* corresponds to the lognormal distribution of particles diameters:

$$f(D) = \frac{1}{\sqrt{2\pi} SD} \exp\left(-\frac{\ln^2 (D / D_0)}{2S^2} \right) \tag{2}$$

with parameters D_0 defined by $\ln D_0$ as the mean of *f(D)* and *S* the mean deviation of *f(D)* from its mean value, known to properly describe the particle size distribution in ferrofluids [17,18].

The magneto-granulometric analysis for the ferrofluid sample performed at temperature *T=300K,* where the Langevin model is applicable is based on the hypothesis of non-interacting particles [16,17], according to which:

$$D_0^3 = \frac{6 k_B T}{\mu_0 \pi H_0 M_s} \sqrt{\frac{M_{sat}}{3 \chi_i H_0}} \tag{3}$$

$$S = \frac{1}{3} \sqrt{\ln \frac{3 \chi_i H_0}{M_{sat}}} \tag{4}$$

$$n = \frac{\mu_0 H_0 M_{sat}}{k_B T} \tag{5}$$

where M_s is the bulk magnetite saturation magnetization. The mean particle diameter D_m was computed using:

$$D_m = D_0 \exp\left(\frac{S^2}{2} \right). \tag{6}$$

The value of initial susceptibility is obtained from the slope of the curve M versus H in the neighborhood of the origin (Fig. 2) and the values of H_0 and M_{sat} result from the extrapolation of the linear part of the curve in Fig. 3.

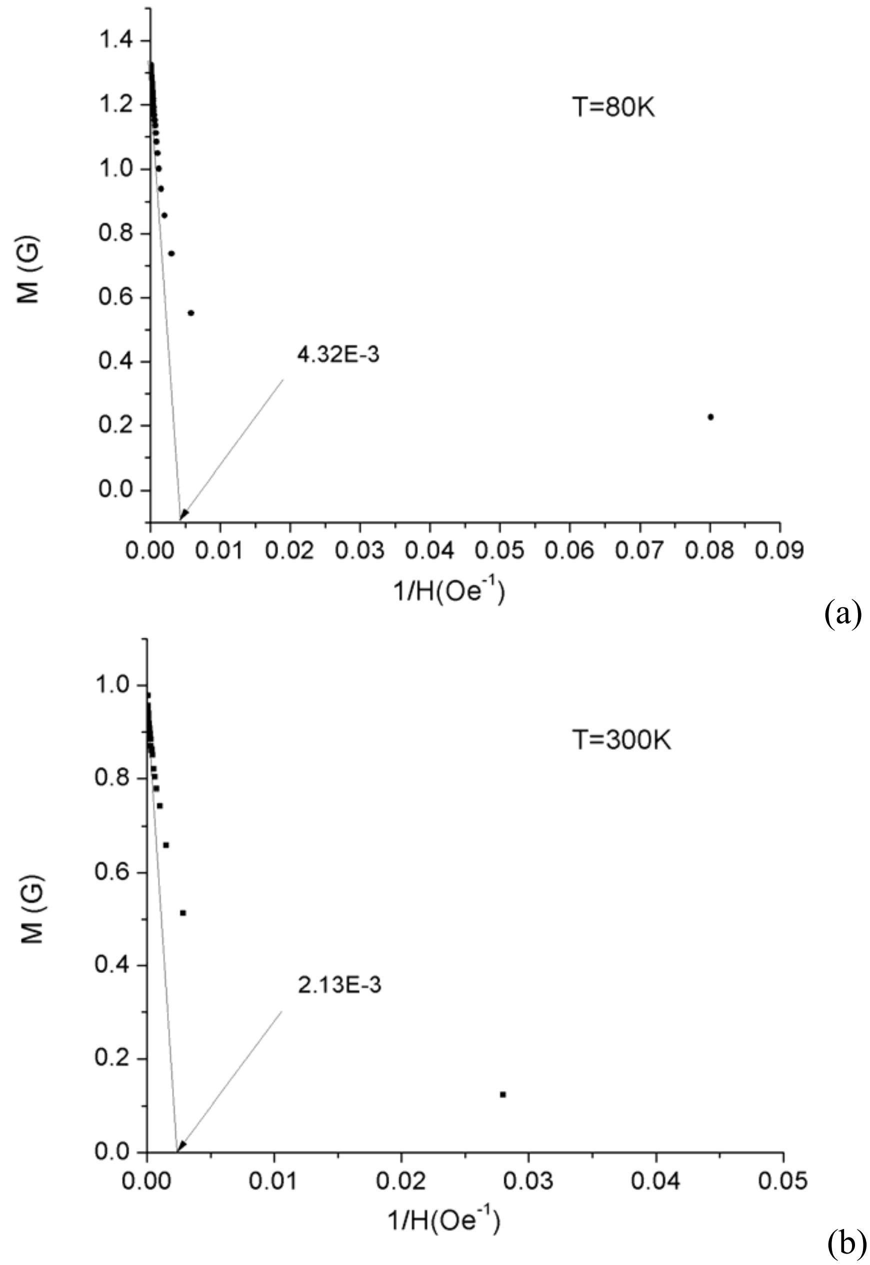

Fig.3 Dependence of the magnetization versus *1/H* for T=80K (a) and T=300K (b)

Figures 3(a) and (b) show the dependences of magnetization values versus $1/H$ in the two cases of extreme temperature. The values represented are restricted to the interest zone of abrupt decreasing of the magnetization. The slope of the linear parts in the two cases are ploted and the exact value for $1/H_0$ in each graph is marked. The numerical values are given in Table 1.

The experimental results obtained for T=80K cannot be treated using the Langevin model. On the assumption that the saturation magnetization is a consequence of the effect of a total orientation of the all individual magnetic moments in the direction of the external field, we can use the relation:

$$M_{sat,80} = \frac{n\pi D_{m.80}^3}{6} M_{s,80}\,. \tag{7}$$

Assuming the condition that the density number of particles does not change we can compute the mean diameter using the values obtained at 300K [19]:

$$D_{m,80} = D_{m,300}\left(\frac{M_{s,300}}{M_{s,80}}\cdot\frac{M_{sat,80}}{M_{sat,300}}\right)^{1/3}. \tag{8}$$

The value for the ratio $\dfrac{M_{s,300}}{M_{s,80}}$ was computed using the data given in [20].

The granulometric parameters for this temperature are also given in Table 1.

Table 1. Magneto-granulometry analysis results of the ferrofluid sample

T(K)	M_{sat} (G)	$1/H_0$ (Oe^{-1})	χ_i	S	D_0 (nm)	D_m (nm)
80	1.33	$4.32\cdot10^{-3}$	$2.16\cdot10^{-3}$	0.13	7.45	7.52
300	0.96	$2.13\cdot10^{-3}$	$1.09\cdot10^{-3}$	0.23	6.67	6.85

Using the parameters of the lognormal distribution for particle magnetic diameter (S,D_0) we can reconstruct the corresponding function as in Fig.4. One can observe the differences between the ferrofluids mean diameter and probability values at the two limiting temperatures.

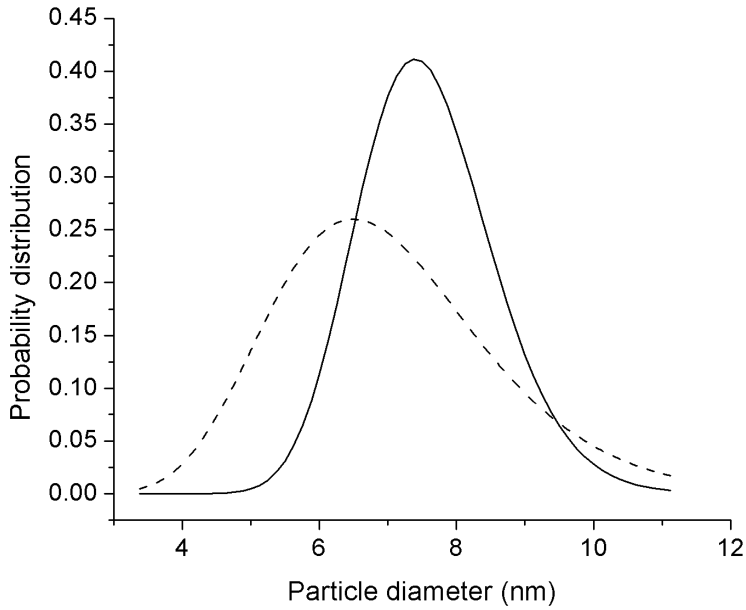

Fig.4 Probability distribution function for particle diameters: T=80K (continuous line) and T=300K (dash line)

4. Conclusion

The results of the experimental investigations demonstrate that the saturation magnetization is higher at lower temperature. Based on the experimental magnetization curve at 300K, using the Langevin model and the hypothesis of lognormal distribution of the particle diameter we compute the parameters of the distribution and construct the probability distribution function. We extend the analysis in the case of 80K where the Langevin model is not valid and obtain the corresponding lognormal distribution.

The comparison of the two distributions shows an increasing in the value of the mean magnetic diameter and a significant diminishing in the spread of the diameters at the lower temperature.

This effect might be a consequence of the increase of the mean volume of the ferromagnetic core of nanoparticles with the decreasing of the temperature. Our experimental result, similar to those presented in [19], seems to be consistent with the "core-shell" model in which at the surface of nanoparticles there exists at room temperature a paramagnetic layer [7,8,10,12,18] becoming gradually ferrimagnetically ordered as the temperature decreases. The consequence of this change might be the increase of the mean volume of the magnetic core of the nanoparticles with the decrease of the temperature.

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