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ASSESSMENTS FOR THE COUPLING OF SUPERPARAMAGNETIC PARTICLES

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Abstract: *The physical analytical conditions for coupling of superparamagnetic particles have been regarded in the present paper. The cases of two interacting superparamagnetic particles and superparamagnetic particle with another one being in single domain state were investigated. Assessments have been made for the critical period of magnetic moments’ oscillations, determining the coupling of superparamagnetic particles. Some conclusions about the role of external magnetic field in this coupling were outlined.*

Keywords: *tribological technology, ferrofluids, superparamagnetic particles, magnetostatics interactions, coupling*

1. INTRODUCTION

The quality of ferrofluids, composing the wide basics of tribology, as a colloidal solution with fine ferro- and ferrimagnetic particles of 1-10 nm linear dimensions by the order of magnitude, is sufficiently dependent on the type of these particles and their interactions [1,2]. These particles normally are in superparamagnetic state and their agglomeration in clusters, chains, couples under the magnetostatics interactions depends both on material micro structure, magnetic properties, geometrical parameters and environmental factors such as temperature and external magnetic field. Thermal activation creates two main types of magnetic moment oscillations and rotations in SP particles. One is called internal, stipulated by the thermal fluctuations, another is called external, stipulated by the rotation of a particle as a whole [1].

As far as the energetical barriers for the in-phase and antiphase magnetic moments’ fluctuations are different of each other the physical conditions for the agglomeration turn up different as well [3].

The physical conditions for the agglomeration of superparamagnetic (SP) particles through the magnetostatics interaction is under the consideration in the present work regarding in-phase and antiphase thermal activation fluctuation modes of their magnetic moments.

2. OBJECTIVES

The objective of the present study is to find out the critical conditions for the agglomeration of superparamagnetic particles with taking into account of in-phase and antiphase modes of thermally activated fluctuations of their magnetic moments.

The knowledge about these conditions is supposed to be important for the further progress in designing of high quality ferrofluids for the tribological technical and technological applications.

2.1 Methods

To analyze the SP particles' magnetostatics interactions the states of these particles, their geometrical and physical parameters are taken into account according to the classical representations [1,2,4].

As far as the shape of the particles under the present study is assumed for the simplicity to be spherical, the magnetostatics interaction between them is the same as for the magnetic dipoles. Two superparamagnetic particles with spherical shape, interacting magnetostatically, are under the consideration in the present study (see figure 1). Influence of magneto crystalline anisotropy is to be taken into account additionally.

The free energy of magnetic dipole interaction can be emphasized as the following:

$$W = \frac{\overrightarrow{\mu_1} \cdot \overrightarrow{\mu_2}}{|\overrightarrow{d}|^3} - \frac{3(\overrightarrow{\mu_1} \cdot \vec{d})(\overrightarrow{\mu_2} \cdot \vec{d})}{|\overrightarrow{d}|^5}, \qquad (1)$$

where vectors $\overrightarrow{\mu_1}$ and $\overrightarrow{\mu_2}$ are the particles' magnetic moments, $\vec{d}$ is the vector, connecting their centers in the right angular coordinate system $oxyz$ [1,4].

By means of writing down the components of all vectors in the formula (1) in the system $oxyz$ one can take the specific relations for the interaction energies for the cases of in-phase and antiphase rotations of the magnetic moments, emphasized in terms of the deflection angle from the initial equilibrium state.

For the analysis of the SP particles' movement the classical mechanics laws is applied with taking into account the interaction (1). For obtaining the results the relevant differential equation of the second order is under the solution with taking into account of the initial conditions.

3. COUPLING OF TWO SP PARTCILES

Couples of SP particles with in-phase and antiphase modes can be regarded as two interacting magnetic dipoles (figure 1). Magnetic moments in the coordinate system $oxyz$ for the in-phase mode can be represented in components as

$$\vec{\mu}_1\{\mu \sin\theta; 0; \mu\cos\theta\},$$

$$\vec{\mu}_2\{\mu \sin\theta; 0; \mu\cos\theta\},$$

where μ is the modulus of the moments $\overrightarrow{\mu_1}$ and $\overrightarrow{\mu_2}$, θ is the deflection angle $\vec{\mu}$ from oz axis. For the antiphase mode –

$$\vec{\mu}_1\{-\mu \sin\theta; 0; \mu\cos\theta\},$$

$$\vec{\mu}_2\{\mu \sin\theta; 0; \mu\cos\theta\}.$$

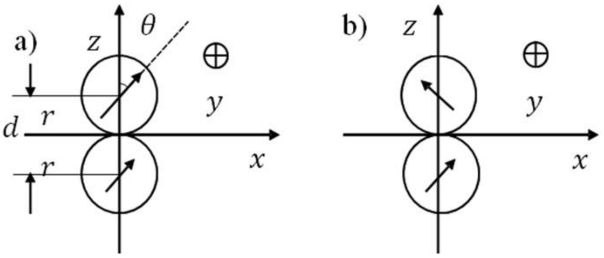

Figure 1. SP particles with in-phase (a) and antiphase (b) magnetic moments

Taking in mind that $\vec{d}$ has the only non-zero component d in oz one can write down the energy (1) for the in-phase and antiphase modes accordingly as

$$W_\mathrm{a} = W_0(1 - 3\cos^2\theta), \qquad (2)$$

$$W_\mathrm{b} = -W_0(1 + \cos^2\theta), \qquad (3)$$

where W_0 is the reference energy, equal to μ^2/d^3.

The energy barriers for these modes are represented in the figure 2.

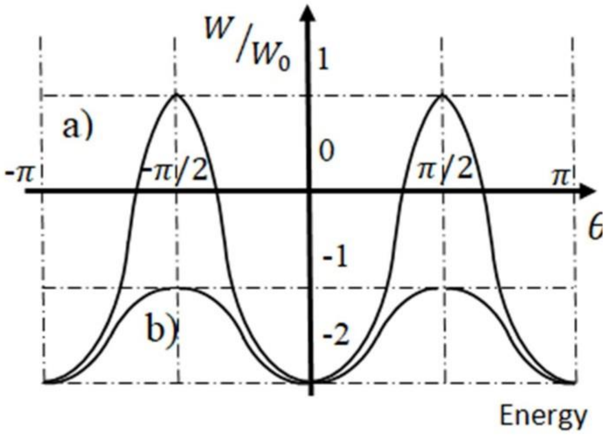

Figure 2. Energy barriers for in-phase (a) and antiphase (b) magnetic moments modes

As far as the particles with antiphase magnetic moments mode turn up always in the negative energy barrier the conditions for their agglomeration doesn't break and they remain in a coupling state. The particles with in-phase mode at the deflection angles θ (3) larger than $\theta_0 = \arccos \sqrt{3}/_3$ and less than $\pi - \theta_0$ repel each other and therefore they can be principally disintegrated if the divergence distance between them will be sufficiently large.

According to the classical mechanics one can write the following equation for the movement of a particle in the time t taking in mind (3)

$$\frac{\mathrm{d}^2 z}{\mathrm{d}t^2} = -\frac{3\mu^2}{md^4} \cdot (1 + 3\cos 2\theta), \qquad (4)$$

where m is the mass of a particle and θ is oscillating with the round frequency ω: $\theta = \omega t$.

Equation (4) shows that the character the particles do move is dependent both on their magnetic interactions and inertia factor.

To find the conditions for the remaining particles' coupling state let's assume for simplicity that they don't move far from each other so that the divergence distance $\Delta d \ll 2r$.

4. CALCULATIONS

By solving the differential equation (4) under the initial conditions at $t = t_0$, when $\theta = \theta_0$, so that

$$z = r; \ \frac{\mathrm{d}z}{\mathrm{d}t} = 0,$$

we find the critical term for the frequency ω and relevant period of magnetic moment oscillations:

$$\tau_{cr} \cong 10 \cdot \sqrt{\rho \cdot \frac{r}{J}}, \qquad (5)$$

where ρ is the particles' material density.

For instance, for the particles with J = 500 Gs, $r = 10^{-6}$ cm, $\rho = 4\ \mathrm{g/cm^3}$ we obtain τ_{cr}= $4{\cdot}10^{-8}$ c. If to compare this value with the internal relaxation time for SP particles, equal to 10^{-9} c [1], we can conclude, that for this kind of thermal activation the agglomeration is stable. According to the term (5) the agglomeration and coupling of the SP particles remain if the frequency of magnetic moments rotation is enough high, i.e. the period is smaller than τ_{cr}. The physical meaning of the relation (5) obtained signifies that the agglomeration of a couple of SP particles for in-phase magnetic moments' oscillations will be realized for the particles with high enough inertia, i.e. having large density and volume impact, low enough magnetization, providing moderate repulsion between them. Otherwise, the particles detach on sufficient distance from each other and the probability for the restoring coupling and agglomeration has low probability, because of collisions with other particles and fluid flows carrying them away.

The energy barriers in SP particles besides of magnetostatics interactions is created also by magneto crystalline anisotropy [1,2,4]. Let's compare the impacts of magnetostatics interaction (1) and magnetic anisotropy free energies. From (1) and (2) for the maximum magnetostatics energy barrier we have

$$W_m = \frac{\pi J^2 V}{3}$$

and for the single axial magneto crystalline anisotropy –

$$W_K = KV,$$

where K is magneto crystalline anisotropy constant and V is a particle's volume.

Comparing the last equalities, we find that magnetostatics interactions prevail, if the following nonequality is fulfilling:

$$\pi J^2 > 3K.$$

Some examples for the assessments are in the table. One can notice that for some SP particles the magnetostatics plays more important role in determining the thermal activation of the magnetic moments than magneto crystalline anisotropy.

Table. Examples for the assessments of the magnetostatic and magnetocrystalline anisotropy free energies' impact

No	J, Gs	K, erg/cm^3	W_m / W_K
1	1000	$3{\cdot}10^6$	0.35
2	500	$5{\cdot}10^4$	2.13
3	1700	$4{\cdot}10^5$	7.57

5. COPUING OF SP AND SD PARTICLES

Let’s consider the case, when one of the coupling particles is in single domain (SD) state and another one is SP (figure 3). The magnetic moment of the first is pinned by the magneto crystalline anisotropy.

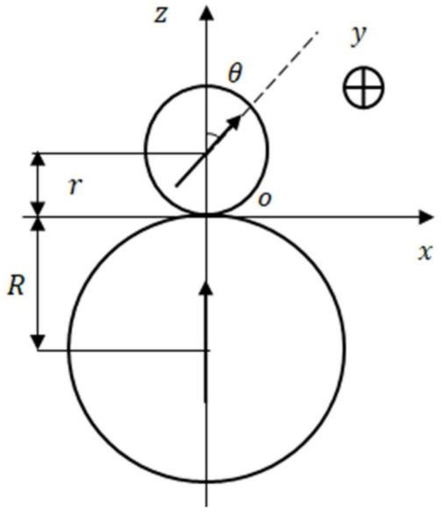

Figure 3. SD and SP particles.

The magnetic moments of the SD and SP partciles in components let’s write accordingly as the following:

$$\vec{\mu}_{SD}\{0{,}0, \mu_{SD}\cos\theta\},$$

$$\vec{\mu}_{SP}\{\mu_{SP}\sin\theta, 0, \mu_{SP}\cos\theta\}.$$

Inserting these components in (1) leads to the impression for the magnetostatics energy of interaction:

$$\boldsymbol{W_1} = \frac{-3\boldsymbol{\mu_{SD}\mu_{SP}}\,\mathbf{cos}\,\boldsymbol{\vartheta}}{(\boldsymbol{r}+\boldsymbol{R})^3}\,. \qquad (\boldsymbol{6})$$

Like for the case of two SP particles by means of composing the equation for the SP particle’s movement (4) one can obtain the condition for the periods of SP particle oscillation, when coupling is conserving:

$$\tau_1 \leq \pi\sqrt{\frac{2\rho r R}{3(\pi-2)J_{SD}J_{SP}}}\,. \qquad (7)$$

where J_{SD},J_{SP} are the magnetizations of the SD and SP partciles accordingly.

Than it follows from (7) that for the smaller SD and SP partciles, larger their magnetizations, more density of SP partcile the periods of oscillations for the coupling should be shorter.

For instance for the magnetizations J_{SP} = 500 Gs, J_{SD} = 1000 Gs, r = 10^{-7} cm, R = 10^{-5} cm, ρ = 4 g/cm^3 (7) gives, that critical period is about $5{\cdot}10^{-9}$c, i.e. five times exceeding the internal relaxation time for SP particles, equal to 10^{-9} c [1].

If to apply external magnetic field, directed along $\boldsymbol{oz}$ axis, the energy barriers for the SP particle’s magnetic moment equilibrium state will be deeper and therefore the average magnetic moment component along this axis will be larger increasing the attraction between SD and SP particles and promoting the coupling between these particles. If to compare the barriers of the magnetostatics energy in an external magnetic field $\boldsymbol{H}$ and single axis magneto crystalline one for the particle’s magnetic moment,

$$\boldsymbol{HJ} \geq \boldsymbol{K},$$

for instance, at $\boldsymbol{J}$ = 500 Gs, $\boldsymbol{K}$ = $5{\cdot}10^5$ erg/cm^3, we can find that the magnetic field, exceeding 100 Oe, plays sufficient role in the coupling of SP particles.

6. CONCLUSION

For the in-phase magnetic moments’ oscillations in SP spherical particles the terms for stable agglomeration of their couples are fulfilling if the particles have enough density, low magnetization and relatively large radius.

The conditions for the SP particles' agglomeration depends also on the acting temperature both trough magnetization getting down and thermal activation rising up.

In some cases the magnetostatics interactions prevail the impact of magneto crystalline anisotropy in magnetic moments’ oscillations in SP particles and in-phase oscillation modes’ are to be taken into the consideration in determining their agglomeration state.

An external magnetic field is able to reduce free energy barrier for the stability of SP partcile’s magnetic moment and therefore increase the probability of the coupling.

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