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Specific Absorption Rate of Assembly of Magnetite Nanoparticles with Cubic Magnetic Anisotropy

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Abstract

The presence of strong magnetic dipole interaction in assemblies of fractal clusters of nearly spherical magnetite nanoparticles, which arise in a biological media loaded with magnetic nanoparticles, leads to a significant decrease of the specific absorption rate of these assemblies in alternating magnetic field. However, the specific absorption rate of the assembly can be increased if the nanoparticles are covered by non magnetic shells of sufficiently large thickness comparable with the nanoparticle diameter.

Keywords: Magnetite nanoparticles, Magneto- dipole interaction, Specific absorption rate, Numerical simulation

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1. Introduction

Superparamagnetic magnetite nanoparticles are used in magnetic nanoparticle hyperthermia [1, 2] for local heating of biological media in low frequency alternating magnetic field. It is well known [3] that perfect magnetite nanoparticles have cubic type of magnetic anisotropy with negative cubic anisotropy constant K_c = - 10^5 erg/cm^3. As a result, the total magnetic energy of such single-domain nanoparticle has 8 equivalent energy minima separated by relatively low energy barriers. This makes these nanoparticles promising for application in biomedicine [4, 5].

In this paper the specific absorption rate (SAR) of assemblies of spherical magnetite nanoparticles with cubic anisotropy is calculated in the range of particle diameters D = 30 - 60 nm, bearing in mind that the single-domain diameter of spherical magnetite nanoparticle equals D_c = 64 nm [6]. The numerical simulations are performed by solving stochastic Landau- Lifshitz (LL) equation [7-9]. This approach takes into account

both thermal fluctuations of the particle magnetic moments and strong magneto-dipole interaction [10-11] in assemblies of dense fractal-like aggregates of nanoparticles. It has been discovered recently [12] that fractal clusters of nanoparticles originate in biological cells loaded with magnetic nanoparticles. Therefore, the influence of magneto- dipole interaction on the assembly behavior is essential.

2. Numerical Simulation

Dynamics of the unit magnetization vector $\vec{\alpha}_i$ of *i*-th single-domain nanoparticle of the cluster is determined by the stochastic Landau- Lifshitz equation [7-9]

$$\frac{\partial \vec{\alpha}_i}{\partial t} = -\gamma_1 \vec{\alpha}_i \times \left(\vec{H}_{ef,i} + \vec{H}_{th,i} \right) - \kappa \gamma_1 \vec{\alpha}_i \times \left(\vec{\alpha}_i \times \left(\vec{H}_{ef,i} + \vec{H}_{th,i} \right) \right), \quad i = 1, 2, ..N_p, \qquad (1)$$

where γ is the gyromagnetic ratio, κ is phenomenological damping parameter, $\gamma_1 = \gamma/(1+\kappa^2)$, $\vec{H}_{ef,i}$ is the effective magnetic field and $\vec{H}_{th,i}$ is the thermal field. The effective magnetic field acting on a separate nanoparticle can be calculated as a derivative of the total cluster energy

$$\vec{H}_{ef,i} = -\frac{\partial W}{M_s V \partial \vec{\alpha}_i}, \qquad (2)$$

where *V* is the volume of *i*-th nanoparticle, M_s = 450 emu/cm^3 is the saturation magnetization of magnetite.

The total magnetic energy of the cluster $W = W_a + W_Z + W_m$ is a sum of magnetic anisotropy energy W_a, Zeeman energy W_Z of the particles in applied magnetic field, and the energy of mutual magneto-dipole interaction of the particles W_m. For nanoparticles with cubic type of magnetic anisotropy the magneto-crystalline anisotropy energy is given by

$$W_a = K_A V \sum_{i=1}^{N_p} \left(\left(\vec{\alpha}_i \vec{e}_{1i}\right)^2 \left(\vec{\alpha}_i \vec{e}_{2i}\right)^2 + \left(\vec{\alpha}_i \vec{e}_{1i}\right)^2 \left(\vec{\alpha}_i \vec{e}_{3i}\right)^2 + \left(\vec{\alpha}_i \vec{e}_{2i}\right)^2 \left(\vec{\alpha}_i \vec{e}_{3i}\right)^2 \right). \qquad (3)$$

Here ($\mathbf{e}_{1i}$, $\mathbf{e}_{2i}$, $\mathbf{e}_{3i}$) is a set of orthogonal unit vectors that determine space orientation of *i*-th nanoparticle of the cluster.

Zeeman energy of the cluster in applied alternating magnetic field $\vec{H}_0 \sin(\omega t)$ is given by

$$W_Z = -M_s V \sum_{i=1}^{N_p} \left(\vec{\alpha}_i \vec{H}_0 \sin(\omega t) \right). \qquad (4)$$

Next, for nearly spherical uniformly magnetized nanoparticles the magnetostatic energy of the cluster can be represented as the energy of the point interacting dipoles

located at the particle centers $\mathbf{r}_i$ within the cluster. Then the energy of magneto-dipole interaction is

$$W_m = \frac{M_s^2 V^2}{2} \sum_{i \neq j} \frac{\vec{\alpha}_i \vec{\alpha}_j - 3\left(\vec{\alpha}_i \vec{n}_{ij}\right)\left(\vec{\alpha}_j \vec{n}_{ij}\right)}{\left|\vec{r}_i - \vec{r}_j\right|^3}, \tag{5}$$

where $\mathbf{n}_{ij}$ is the unit vector along the line connecting the centers of i-th and j-th particles, respectively.

The thermal fields $\vec{H}_{th,i}$ acting on various nanoparticles of the cluster are statistically independent, with the following statistical properties [13] of their components

$$\left\langle H_{th.i}^{(\alpha)}(t) \right\rangle = 0; \left\langle H_{th,i}^{(\alpha)}(t)\, H_{th,i}^{(\beta)}\left(t_1\right) \right\rangle = \frac{2k_B T\kappa}{\gamma M_s V} \delta_{\alpha\beta} \delta\left(t - t_1\right), \alpha, \beta = (x, y, z). \tag{6}$$

Here k_B is the Boltzmann constant, $\delta_{\alpha\beta}$ is the Kroneker symbol, and $\delta(t)$ is the delta function.

The geometry of a fractal cluster of single-domain nanoparticles is characterized [14] by the fractal descriptors D_f and k_f. By definition, the total number of nanoparticles N_p in a fractal cluster equals $N_p = k_f \left(2R_g/D\right)^{D_f}$, where D_f is the fractal dimension, k_f is the prefactor, R_g being the radius of gyration. The fractal clusters with various fractal descriptors were created in this paper using the well known Filippov's et al. algorithm [14]. The calculations were performed for fractal clusters with fractal descriptors D_f = 1.9 and k_f = 1.7 because these clusters are most often observed in biological cells [12] loaded with magnetic nanoparticles.

3. Results and Discussion

Fig. 1a shows the geometrical structure of a fractal cluster with fractal descriptors D_f = 1.9 and k_f = 1.7 consisting of N_p = 100 spherical nanoparticles. One can see that in a fractal cluster considerable number of the nanoparticles turns out to be at closest distances from each other. It is supposed that each nanoparticle of diameter D is covered by a non magnetic shell of thickness t_{sh}, as Fig. 1b shows. The non magnetic shells protect magnetite nanoparticles from further oxidation. They prevent also direct exchange interaction between neighbouring nanoparticles of the cluster [4, 5].

The low frequency hysteresis loops of dilute assemblies of random fractal-like clusters consisting of N_p = 100 spherical magnetite nanoparticles have been calculated using stochastic Landau- Lifshitz equation (1), (2) at a room temperature, T = 300 K, for typical frequency of the alternating magnetic field f = 300 kHz and in the range of magnetic field amplitudes H_0 = 50 - 100 Oe. Then the SAR of the assemblies was calculated [11] as $SAR = 10^{-7} M_s f A/\rho$ (W/g), where A is the hysteresis loop area in the

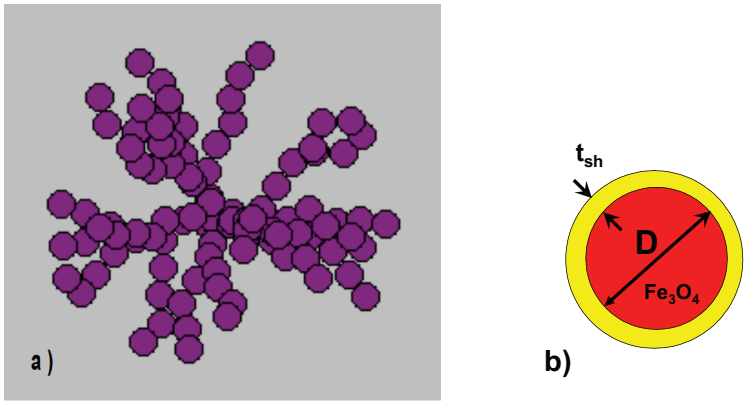

Figure 1: a) Geometry of fractal cluster of single-domain nanoparticles with fractal dimension D_f = 1.9 and prefactor k_f = 1.7; b) spherical magnetite nanoparticle of diameter D covered by a non magnetic shell of thickness t_{sh}.

variables (M/M_s, H), ρ = 5 g/cm^3 being the density of magnetite nanoparticles. To see clearly the influence of the magneto- dipole interaction on the SAR value, we calculate also the low frequency hysteresis loops for assemblies of non interacting magnetite nanoparticles of various diameters.

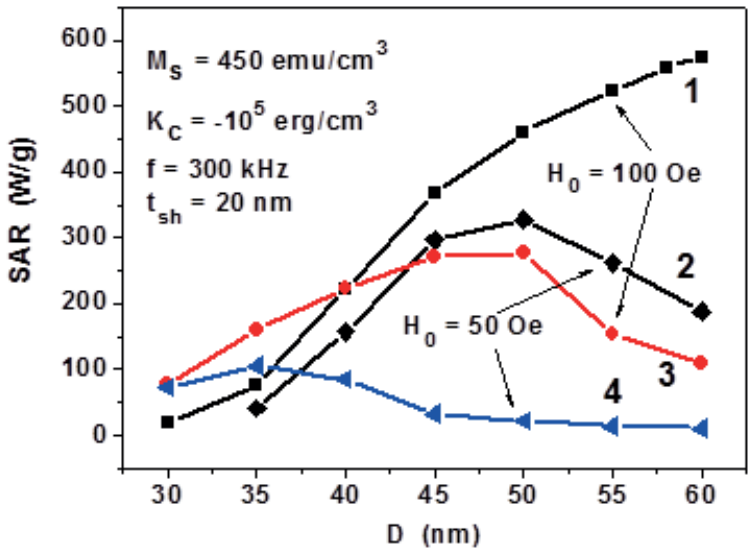

Figure 2: SAR of assembly of magnetic nanoparticles with cubic anisotropy as a function of particle diameter at various amplitudes of alternating magnetic field H_0: 1), 2) – assemblies of non interacting nanoparticles, 3), 4) dilute assemblies of fractal clusters with strong magneto-dipole interaction.

As Fig. 2 shows, at frequency f = 300 kHz SAR of the non interacting nanoparticle assemblies (curves 1) and 2) in Fig. 2) can reach large values, on the order of 300-500 W/g. However, the presence of strong magnetic dipole interaction between the nanoparticles of fractal clusters leads to a significant decrease of the assembly SAR (curves 3), 4) in Fig. 2). One can see in Fig. 2, that at the magnetic field amplitude H_0 = 100 Oe in the range of the optimal particle diameters, D = 45-55 nm, the SAR

of the assembly of fractal clusters decreases by 30-40% compared to that of a non interacting nanoparticle assembly.

The results shown in Fig. 2 correspond to assemblies of nanoparticles of various diameters, but having the same thickness of the non magnetic shells, t_{sh} = 20 nm, at the nanoparticle surfaces. It is found however, that the shape of the low frequency hysteresis loops, as well as the SAR, depends considerably on the non magnetic shell thickness.

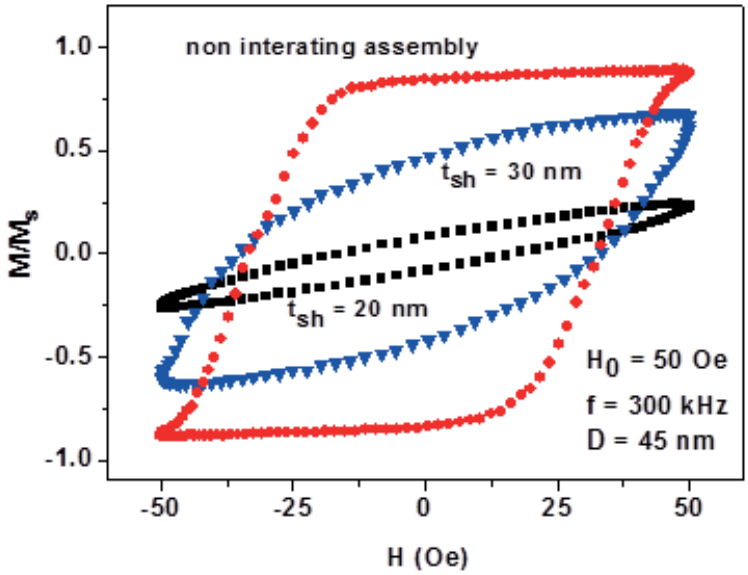

Figure 3: Low frequency hysteresis loops of dilute assemblies of fractal clusters consisting of N_p = 100 spherical magnetite nanoparticles of the same diameter D = 45 nm, but having different non magnetic shell thickness.

As Fig. 3 shows, for a particular particle diameter, D = 45 nm, the hysteresis loop of the assembly of nanoparticles with t_{sh} = 20 nm has the lowest area. The area of the hysteresis loop increases considerably for nanoparticles with t_{sh} = 30 nm, however the hysteresis loop of the largest area corresponds to non interaction assembly of magnetite nanoparticles. Evidently, the distance between the magnetic cores of the closest nanoparticles in the fractal cluster increases as a function of t_{sh}. This leads to a decrease in the intensity of the magneto-dipole interaction among the nanoparticles of the cluster. As a result, the SAR of the assembly of fractal clusters increases.

4. Summary

In this paper the low frequency hysteresis loops of dilute assemblies of fractal clusters of spherical magnetite nanoparticles have been calculated using the stochastic Landau- Lifshitz equation. The calculations have been carried out for the range of the

nanoparticle diameters D=30-60 nm for various thicknesses t_{sh} of the non magnetic shells at the nanoparticle surfaces. It is found that for clusters of interacting nanoparticles the SAR of the assembly depends appreciably on the average particle diameter and raises with the non magnetic shells thickness increase. The highest SAR is obtained for the assemblies of non interacting magnetite nanoparticles.

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