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ENHANCE HEATING EFFICIENCY OF MAGNETIC NANOPARTICLES

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Abstract

The use of magnetic nanoparticles for cancer therapy requires an appreciable heating power generated by the applied magnetic field. Therefore, it is of great importance to enhance the efficiency of the heating mechanism for example by determining an ideal choice of input parameters (e.g. material properties). Another way to increase the heat generation is related to the applied field. We show that a rotating applied field, (as distinct from the oscillating one) in magnetic nanoparticles with uniaxial anisotropy, can produce for us a more efficient heating. This is found in calculations of the energy loss per cycle previous to the steady state known as the long-time solution of the Landau-Lifshitz-Gilbert equation.

I. Introduction

Magnetic nanoparticles have numerous medical applications, of which we are concerned with hyperthermia, heat treatment. The unique feature of magnetic nanoparticle hyperthermia is that the energy is transported in the body by means of an ac magnetic field. The nanoparticles absorb the energy and turn it in heat. The magnetic moment of the particles enable also targeting: they can be directed towards the cancer tumors by a magnetic field.

Hyperthermia therapy for cancer is a medical treatment in which tissue temperatures are elevated for the purpose of damaging or destroying cancer cells. A sustained temperature between 41 °C and 45 °C can cause irreversible damage to cell function, which predominantly lead to preprogrammed cell death, know as apoptosis. Apoptosis is form of intentional cell death based on a genetic mechanism. A sustained temperature above 45 °C causes another form of cell death, commonly referred to as necrosis, the general term for the path to cell death.

Although hyperthermic cancer treatment is able to heat the tumor region to apoptotic temperatures (which lead to apoptotic cell death) but it is used mainly together with chemotherapy or radiotherapy. Since particular tumor cells are sensible for heating, magnetic nanoparticles can be used for hyperthermia which receives important applications in cancer therapy especially when the ordinary treatments are not applicable. For instance brain tumors are among the most difficult forms of cancer to treat, although fever therapy is know to be very effective. But fever therapy is a form of whole-body hyperthermia. Limiting the heat to the brain would be desirable, but that would require very accurate controle of temperature. Concerns of this nature delay the wide introduction of magnetic nanoparticle hyperthermia in cancer treatment. Hence, the study of relaxation mechanisms of magnetic nanoparticles is a very active research field both in the theoretical and material-science aspects.

The research to be reported below is part of the studies on the relaxation of magnetic nanoparticle systems under oscillating and rotating polarization of the applied field. A theoretical investigation of the isotropic, single-particle case has been done and the results have been published [1].

This work was based on the Landau-Lifshitz-Gilbert (LLG) equation which has been widely used to investigate the nonlinear dynamics of magnetisation and the specific power loss of magnetic nanoparticles systems and it was followed by a similar work based on the modified Bloch equation [2]. Apart from following the response of the magnetization of nanoparticles to the applied ac field, the frequency dependence of the energy loss (i.e. the heat gain) was determined. For both models, is was found in the low frequency limit, the energy loss per cycle is larger in oscillating applied field than in rotating field. However, the comparison to experimental data [3] requires the generalization of the previously obtained results to the anisotropic case. The purpose of our recent paper [4] was to determine the effect of anisotropy on the heat production of magnetic nanoparticles under rotating field. Numerical calculations on the LLG equation have shown that the power loss decreases under the anisotropy. The outcome of our research was that in the low frequency limit in rotating field it is impossible to increase the energy loss by means of inserting an anisotropic term into the potential energy into the LLG Eq.(1).

Here we will show that a rotating applied field, (unlike an oscillating field) in uniaxially anisotropic magnetic nanoparticles can produce us a more efficient heating if the energy loss per cycle is calculated outside the steady state of the solution of the Gilbert (or equivalently the LLG) equation.

II. Landau-Lifshitz-Gilbert equation

Out of the many phenomenological equations of motion for the relaxation of magnetization [5] the Gilbert equation [6] has proved to give the most realistic description of the dynamics of single-domain magnetic particles at strong damping. Such a particle, being too small to accommodate a domain wall, can be fully characterized with a single vector, its magnetic moment **m**. An important feature of Larmor precession is that the magnitude of m does not change under the influence of the external field, including the anisotropy field. Hence it is convenient to rewrite the equation of motion of the magnetization **m** of a single-domain particle in terms of the unit vector $\mathbf{M} = \mathbf{m}/m_S$, m_S being the saturation magnetic moment. Then the Gilbert equation reads as

$$\frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M} = \gamma_0 \mathbf{M} \times \left[\nabla V + \mu_0 \eta \frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M} \right], \tag{1}$$

where $\gamma_0 = 1.76 \times 10^{11}$ $\mathrm{Am^2/Js}$ is the gyromagnetic ratio of the electron spin (with opposite sign), $\mu_0 = 4\pi \times 10^{-7}$ Tm/A (or $\mathrm{N/A^2}$) is the permeability of free space, V is the potential energy and η is the damping factor, both of them normalized for unit M. To describe the system, the potential energy must contain the Zeeman energy in the magnetic field and the anisotropy energy $(\mu_0/2)MH_a\sin^2(\theta)$ [7]. We define the vector $\mathbf{H}$, which contains the external applied magnetic field (here we chose a circularly polarized one) and the effect of the anisotropy of the magnetic particle

$$\mathbf{H} = H_0\ (\cos(\omega t), \sin(\omega t), \lambda_{\text{eff}} M_z), \tag{2}$$

where ω is the angular frequency of the applied field, M_z is the z-component of the normalized magnetization vector and $\lambda_{\text{eff}} = H_a/H_0$ is the measure of the strength of the anisotropy field H_a with respect of the applied H_0 field.

The Gilbert equation can be rewritten in such a way that it has a functional form similar to the Landau-Lifshitz equation. This is called the Landau-Lifshitz-Gilbert (LLG) equation,

$$\frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M} = -\gamma'[\mathbf{M} \times \mathbf{H}] + \alpha'[[\mathbf{M} \times \mathbf{H}] \times \mathbf{M}], \tag{3}$$

where $\gamma' = \mu_0\gamma_0/(1+\alpha^2)$ and $\alpha' = \gamma'\alpha$ with the dimensionless damping factor $\alpha = \mu_0\gamma_0\eta m_S$.

The magnitude of the magnetization vector being constant, the M component of the Gilbert equation (or equivalently the LLG equation (3)) is useless. It can be shown that the equations of motion of the θ and φ components of the unit magnetization are

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M}_\theta &= -\omega'_L \sin(\varphi - \omega t) - \alpha\frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M}_\varphi, \\ \frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M}_\varphi &= -\omega'_L \cos\theta\cos(\varphi - \omega t) + \omega_a \sin\theta\cos\theta + \alpha\frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M}_\theta, \end{aligned} \tag{4}$$

Here we have introduced, the Larmor frequency $\omega'_L = \mu_0\gamma_0 H_0$ and an analogous frequency parameter related to the anisotropy field previously used by Denisov et al. [6] $\omega_a = \mu_0\gamma_0 H_a$ (note that $\gamma_0 > 0$). By using

$$\frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M}_\theta = \frac{\mathrm{d}}{\mathrm{dt}}\theta, \qquad \frac{\mathrm{d}}{\mathrm{dt}}\mathbf{M}_\varphi = \sin(\theta)\frac{\mathrm{d}}{\mathrm{dt}}\varphi \tag{5}$$

the equations of motion of the polar coordinates reads as

$$\begin{aligned}
\frac{\mathrm{d}\theta}{\mathrm{dt}} &= \omega_L \sin\phi + \alpha_N \cos\theta\cos\phi - \alpha_N\lambda_{\mathrm{eff}}\sin\theta\cos\theta, \\
\frac{\mathrm{d}\phi}{\mathrm{dt}} &= \omega_L \cos\phi\frac{\cos\theta}{\sin\theta} + \omega - \alpha_N\frac{\sin\phi}{\sin\theta} - \omega_L\lambda_{\mathrm{eff}}\cos\theta \qquad (6)
\end{aligned}$$

where $\omega_L = H_0\gamma' = \omega'_L/(1+\alpha'^2)$, $\lambda_{\mathrm{eff}} = \omega_a/\omega'_L$ and $\alpha_N = H_0\alpha' = \alpha\omega_L$. Here, ωt, the circular motion of the magnetic field, is subtracted from the azimuthal φ, leaving the lag of $\mathbf{M}$ behind $\mathbf{H}$. As $\mathbf{M}$ rotates behind $\mathbf{H}$, we introduce $\phi = (\omega t - \varphi)$ as the measure of lagging. The latest definition shows that the Larmor term in both equations under (6) is dominating over the second term, because α', Landau-Lifshitz's "dimensionless damping constant", is supposed to be small ($\ll 1$). On the other hand, the last term may be the overdog in both equations. The anisotropy field of magnetite is more than 40 kA/m, and if we reckon with the shape anisotropy as well, as suggested by Bertotti et al. [7], a much stronger effect can be expected.

III. Steady state solution of the LLG equation

The solution of Eq. (4) (which is derived from the Gilbert or the equivalent LLG equation) is shown in Fig. 1 (for the construction of orbit maps and the appearance of steady states therein see [10]). The set of parameters used in the figure are listed in the caption. Here λ_{eff} is closely below the critical value, beyond which there are two attractive fixed points. The attractive fixed point of Fig. 1 corresponds to a steady state solution of the original (unrotated) Gilbert or LLG equation,

$$\begin{aligned}
M_x(t) &= u_{x0}\cos(\omega t) - u_{y0}\sin(\omega t), \\
M_y(t) &= u_{x0}\sin(\omega t) + u_{y0}\cos(\omega t), \\
M_z(t) &= u_{z0}. \qquad (7)
\end{aligned}$$

where u_{x0} and u_{y0} are determined by ω, ω_L, α_N and λ_{eff}. The loss energy per cycle is calculated by determining these attractive fixed point solutions. Then the energy dissipated in a single cycle is given as

$$E = \mu_0 m_S \int_0^{\frac{2\pi}{\omega}} \mathrm{dt}\left(\mathbf{H}\cdot\frac{\mathrm{d}\mathbf{M}}{\mathrm{dt}}\right) = \mu_0 2\pi \mathrm{m_S H(-u_{y0})}, \qquad (8)$$

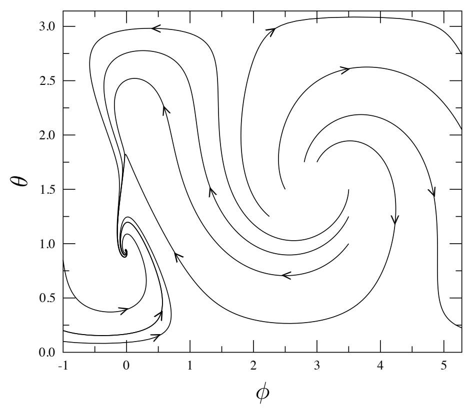

Figure 1: Orbit map in the rotating frame obtained by solving the LLG equation, slightly below the critical value of anisotropy. The parameters are $\alpha_N = 0.1$, $\omega = -0.01$, $\omega_L = 0.2$ and $\lambda_{\text{eff}} = 1.175$. One finds a similar orbit map by changing the direction of the rotating external field ($-\omega$), see Fig.1 of [9]. A single attractive (black dot) and a single repulsive fixed points exist below the crtical value of the anisotropy parameter. Another (attractive) fixed point emerges but only with λ_{eff} above the critical anisotropy, however, its "effect" can be seen on the figure since some of the trajectories merge into a single one before reaching the attractive fixed point.

(see also Eq. (12) in [4]) which has the form in the low-frequency limit, $\omega \ll \alpha_N$, and small anisotropy $|\lambda_{\rm eff}| \ll 1$ limits where one finds a single attract fixed point with

$$u_{y0} \approx -\frac{\alpha_N \omega}{\omega_L^2 + \alpha_N^2} + \frac{\alpha_N \omega_L^2 \omega^3}{(\omega_L^2 + \alpha_N^2)^3}(1 + 2\lambda_{\rm eff}). \tag{9}$$

Inserting (9) into the expression of the energy loss per cycle (8) one finds,

$$E = 2\pi\mu_0 m_S H \left[\frac{\alpha_N \omega}{\omega_L^2 + \alpha_N^2} - \frac{\alpha_N \omega_L^2 \omega^3}{(\omega_L^2 + \alpha_N^2)^3}(1 + 2\lambda_{\rm eff})\right]. \tag{10}$$

Let us note that for axial geometrical anisotropy $\lambda_{\rm eff} > 0$ the nanoparticle has a "cigar-shape" which means it is a prolate ellipsoidal particle and for planar geometrical anisotropy $\lambda_{\rm eff} < 0$ it has a "lens-shape" which means it is an oblate ellipsoidal particle. It is clear that for positive (negative) lambda, the energy per cycle is decreased (increased) by the anisotropy but only for relatively large frequencies.

IV. Out of steady state solution of the LLG equation

Previously we argued that if one relies on the steady state solution of the LLG equation, i.e. the fixed point solution of Eq. (6), the energy loss per cylce cannot be increased by the anisotropy (for low frequencies) in case of a rotating external field. However, it is not the case if the energy loss per cycle is calculated out of the steady states. Indeed, it was shown in [10] that if one plots the energy loss per cycle as a function of various starting points on the $(\theta - \phi)$ plane, a 'well' is found on the 3D graphics which corresponds to the attractive fixed point (i.e. it shows that the steady state solution produces us the lowest energy loss per cycle). While the 'hill' of the 3D graphics, where the energy loss is the maximum, is related to initial conditions taken at the repulsive fixed point. Numerical results are shown for small anisotropy, see Fig. 3 of [10].

Here we repeat the same calculation (same as Fig. 3 of [10]) but for large values of the anisotropy parameter (closer to the critical one), see Fig. 2. It is shown that one finds a similar 3D structure for low anisotropy and for closer to the critical one. The only difference is that closer to the critical

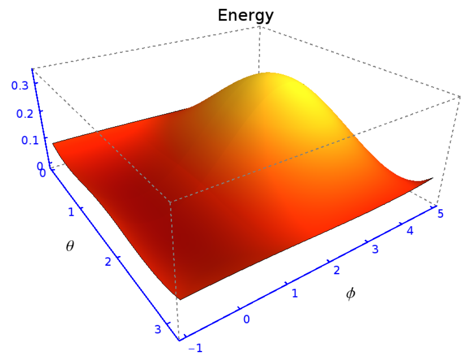

Figure 2: On this 3D graphics one finds the energy loss obtained in the first cycle of the rotating applied field as a function of the initial conditions on the $(\theta - \phi)$ plane. Numerical results are obtained for relatively high anisotropy which is close to (but below) the critical value. The 'hill' corresponds to the largest energy loss found to be situated at the repulsive fixed point. The deepest 'well' is related to the lowest energy loss and found at the attractive fixed point.

value of the anisotropy paremeter two 'wells' appear, but only the lowest is still the one which corresponds to the attractive fixed point (steady state solution). The new 'well' is the consequence of another (attractive) fixed point emerges (only!) above the critical anisotropy, however, its 'effect' can be seen on the Fig. 2 since some of the trajectories merge into a single one before reaching the attractive fixed point and the flow becomes 'slow'.

V. Conclusion

In this work we considered energy losses of magnetic nanoparticles under rotating applied field. The energy loss is calculated in the first cycle of the rotating applied field as a function of the initial conditions on the $(\theta - \phi)$ plane. It is shown that under a rotating applied field, the energy loss per cycle obtained for anisotropic magnetic nanoparticle is maximized if the initial position of the magnetic moment of the particle is chosen to be at the repulsive fixed point of Eq. (6). It is demonstrated that one finds a similar 3D "structure" for low anisotropies and for those closer (but below) to the critical one, i.e. the energy loss has a maximum at the repulsive and a minimum at the attractive fixed points.

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