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Precession of a Fine Magnetic Particle with Finite Anisotropy in a Viscous Fluid

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In this paper, the magnetic dynamics and mechanical rotation about the center of mass of a uniaxial ferromagnetic fine particle in a viscous liquid are described using the classical approach. In particular, the synchronous rotation of the magnetization vector and unit vector, associated with the anisotropy axis, together with a circularly polarized external magnetic field is considered. The feature of this mode for the given system is that both the magnetization and anisotropy axis do not lie in the plane of the field polarization. This fact is explained by the presence of effective permanent field, perpendicular to the polarization plane. Obtained results allow to perform more realistic evaluations of the power loss of an external field interacting with a ferromagnetic fluid.

Keywords: Fine magnetic particle, Equation of motion, Precession, Effective field.

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1. INTRODUCTION

The problem of describing the trajectories of ferromagnetic nanoparticles suspended in a liquid is directly related to the problem of the microscopic description of the response of a ferromagnetic liquid [1] to an external field. First, the state of motion of each particle eventually determines a certain form of their collective behavior which in itself allows to claim analytical results. Second, attention to the microscopic behavior makes it possible to improve approaches to the numerical description of the behavior of ferrofluids and reduce computational costs to obtain the desired result. At the same time, the assigned problem is far from being trivial and has been studied in various approximations for a long time.

The dynamic approximation, in which thermal fluctuations are considered negligible, is important from both the methodological and practical points of view. Thus, we obtain not only the basic idea of the nanoparticle dynamics, but under certain circumstances (the conditions will be discussed below) the results of this approximation are quite adequate to the real situation. The classical equations of motion are the basis for the dynamical description. For example, to describe the precession of the magnetic moment excited by a circularly polarized field in a fixed particle, the Landau-Lifshitz equation was successfully used [2]. For the case of precession in this field of a rigid spherical dipole, the basic equation of rotational motion with a torque of friction proportional to the angular velocity was applied [3].

However, attempts to describe the coupled magnetic and mechanical dynamics of a nanoparticle in a liquid did not always lead to consistent results. Thus, the approximate expressions for the angular characteristics of precession in the case of small field amplitudes were obtained in [4] by linearizing the Lagrange equation. The generalization of the results for the case of arbitrary field frequencies and amplitudes can be performed through the analysis of direct equations of motion. The writing of the latter, as shown by a number of unsuccessful attempts [5, 6], turned out to be a distinctive challenge for theorists. And only relatively recently has a consistent approach been proposed based on the total angular momentum conservation law [7]. Using the equations of motion obtained in the above-mentioned work, we describe the features of precession of the magnetic moment and the easy axis of a nanoparticle with finite anisotropy in a viscous fluid under the action of a rotating field, as well as study the dependences of the characteristics of the stable solutions on the system parameters.

2. THE MODEL AND ITS JUSTIFICATION

We consider a spherical ferromagnetic nanoparticle of radius R, magnetization M and density ρ. The uniaxial anisotropy is characterized by the anisotropy field H_a. We assume that the particle is single-domain and the change in magnetization occurs without changing its magnitude ($|\mathrm{M}| = M = \mathrm{const}$), since all the spin magnetic moments always remain parallel due to the strong exchange interaction. In addition to the motion of the magnetization with respect to the crystal lattice, we suggest that the particle itself can rotate around its center of mass being suspended in a liquid with viscosity η. The translational motion of the particle is not taken into account.

As noted earlier, to describe the coupled magnetic and mechanical rotational dynamics of the nanoparticle, it is not sufficient to simultaneously use the equations describing the magnetic dynamics of a fixed particle and the motion of a particle, whose magnetic moment is rigidly fixed to the crystal lattice. A consistent approach to solving this problem was proposed in [7], where the following system of equations was written based on the total angular momentum conservation law:

$$\dot{\mathrm{n}} = \boldsymbol{\omega} \times \mathrm{n}\,, \tag{1}$$

$$J\dot{\boldsymbol{\omega}} = \gamma^{-1} V\dot{\mathrm{M}} + V\mathrm{M} \times \mathrm{H} - 6\eta V\boldsymbol{\omega}\,, \tag{2}$$

$$\dot{\mathrm{M}} = -\gamma\left(\mathrm{M} \times \mathrm{H}_{eff}\right) + \alpha M^{-1}\left[\mathrm{M} \times \left(\dot{\mathrm{M}} - \boldsymbol{\omega} \times \mathrm{M}\right)\right], \tag{3}$$

where n is the unit vector indicating the direction of the anisotropy axis, $\boldsymbol{\omega}$ is the angular velocity of the particle, J ($= 8\pi\rho R^5/15$) is the moment of inertia, γ is the gyromagnetic ratio, H is the external uniform field, V is the particle volume, α is the damping parameter, H_{eff} is the effective magnetic field which takes into account the internal anisotropy field as

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$$\mathrm{H}_{eff} = \mathrm{H} + H_a M^{-1}(\mathrm{Mn})\mathrm{n}, \qquad (4)$$

and, finally, the dot above denotes the time derivative. In fact, equation (1) is the condition of spherical motion of a rigid body. Equation (2) is the basic equation of rotational motion, in the right side of which, in addition to the moment of the friction force and the moment describing the action of the external field, there is a term proportional to the derivative of magnetization arising from the total angular momentum conservation law. It is precisely this that makes up the difference between equation (2) and its analogue, which is often used to describe the rigid dipole rotation. Equation (3) coincides with the Landau-Lifshitz-Gilbert equation up to a term proportional to $\mathrm{M} \times \boldsymbol{\omega} \times \mathrm{M}$, which excludes the component M rotating along with the crystal lattice. Further we assume that the particle is affected by an external circularly polarized homogeneous field in the form

$$\mathrm{H} = \mathrm{e}_x H \cos\Omega t + \mathrm{e}_y H \sigma \sin\Omega t, \qquad (5)$$

where e_x, e_y are the unit vectors of the Cartesian coordinate system, H and Ω are the field amplitude and frequency, respectively, t is the time, and $\sigma (= \pm 1)$ is the factor determining the field polarization direction.

Equations (1)-(3) are written in the so-called dynamic approximation, when thermal fluctuations are not taken into account. This assumption is true if a number of conditions are fulfilled. First, the magnetic energy should be much greater than the thermal one: $\kappa \gg 1$, $\kappa = MHV/k_\mathrm{B}T$, where k_B ($\approx 1.38 \cdot 10^{-16}$ erg/K) is the Boltzmann constant, T is the thermodynamic temperature. Thus, for example, the condition of such a ratio between the magnetic and thermal energies $\kappa \approx 12$ is fulfilled for maghemite particles [8] with an average radius $R = 20$ nm, magnetization $M = 338$ emu, anisotropy field $H_a = 910$ Oe, at a temperature of $T = 311$ K and field amplitude $H = 0.05 \cdot H_a$.

Making demands on the field amplitude or particle size, it is important to synchronize them with the frequency requirements. Even if the condition $\kappa \gg 1$ is met, there are both random deviations of the magnetic moment with respect to the crystal lattice and random changes in the angular coordinates of the entire particle. And if a sufficiently substantial thermal fluctuation is highly probable over the field period, then the dynamic approximation becomes invalid. The relaxation time is the characteristic time, during which the significant thermally induced changes can occur. And, as a consequence, for the mechanical rotation, the minimum field frequency should be determined by the Brownian relaxation time $\Omega_\mathrm{B} = 1/\tau_\mathrm{B} = k_\mathrm{B}T/(3\eta V)$, for switching processes – by the Neel relaxation time $\Omega_\mathrm{N} = 1/\tau_\mathrm{N} = (\pi/\kappa)^{1/2}(2\alpha\gamma H_a)\exp(-\kappa)$ [9]. Then, the resulting requirement for the external field frequency is written as $\Omega \gg \max[\Omega_\mathrm{B}, \Omega_\mathrm{N}]$. For the above maghemite nanoparticles, taking $\alpha = 0.02$ and blood as a viscous carrier with viscosity $\eta = 0.05$ at a temperature of $T = 311$ K, we obtain that $\Omega_\mathrm{B} \approx 8.54 \cdot 10^3$ Hz and $\Omega_\mathrm{N} \approx 7.61 \cdot 10^3$ Hz. Thus, the frequency should be in the range of not less than hundreds of kilohertz $\Omega \gg 10^4$ Hz that is quite acceptable for most ferrofluid applications including magnetic hyperthermia [10].

3. RESULTS AND DISCUSSION

If the precession mode is implemented, the nanoparticle rotates synchronously with the field (5) (see Fig. 1). The stationary solutions of equations (1)-(3) in this case should be sought in the form

$$\varphi = \sigma\Omega t - \varphi_1, \quad \vartheta = \vartheta_1, \qquad (6)$$

$$\phi = \sigma\Omega t - \phi_1, \quad \theta = \theta_1, \qquad (7)$$

where φ, ϑ are the spherical coordinates of the magnetic moment M; ϕ, θ are the spherical coordinates of the easy axis n; ϑ_1, θ_1 are the precession angles of the vectors M and n, respectively; φ_1, ϕ_1 are the lag angles of the vectors M and n, respectively. The absence of motion of m with respect to the crystal lattice will be a feature of the precession mode, and, therefore, the following condition will be true:

$$\dot{\mathrm{M}} - \boldsymbol{\omega} \times \mathrm{M} = 0. \qquad (8)$$

Substituting expressions (8), (3) into (2) and taking into account (4), we obtain

$$J\dot{\boldsymbol{\omega}} = -H_a M^{-1} V (\mathrm{M} \times \mathrm{n})(\mathrm{M} \cdot \mathrm{n}) - 6\eta V \boldsymbol{\omega}. \qquad (9)$$

To simplify the subsequent calculations, we introduce the coordinate system $x''y''z''$, which rotates along with the easy axis, and $\mathrm{n}'' = \mathrm{e}_z$. According to the application in [2], the condition $\dot{\mathbf{n}}'' = \rho\Omega\sin\theta_1 \mathrm{e}_{y''}$ is satisfied in this coordinate system. Let us find the angular velocity $\boldsymbol{\omega}''$ in the double-primed coordinate system by substituting the last expression into equation (1)

$$\mathrm{e}_{y''}\sigma\Omega\sin\theta_1 = \mathrm{e}_{x''}\omega_{y''} - \mathrm{e}_{y''}\omega_{x''} \Rightarrow$$
$$\boldsymbol{\omega}'' = (-\sigma\Omega\sin\theta_1, 0, 0). \qquad (10)$$

Unfortunately, equation (3) cannot be written in the rotating system; therefore, it is possible to use the result of (10) only by transferring it to a laboratory coordinate system. To this end, we introduce the rotation matrices, the origin of which is clear from Fig. 2, as

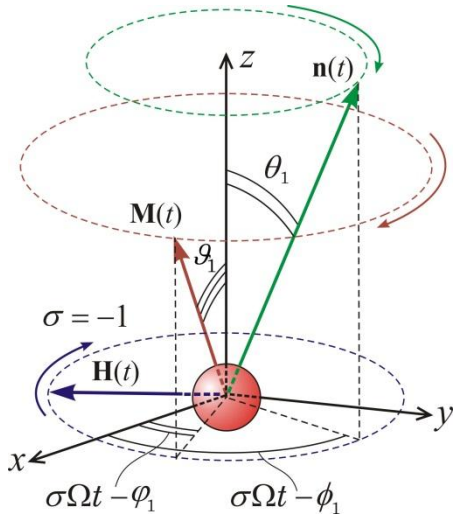

Fig. 1 – Schematic representation of the synchronous rotation of the external field, the nanoparticle and its magnetic moment

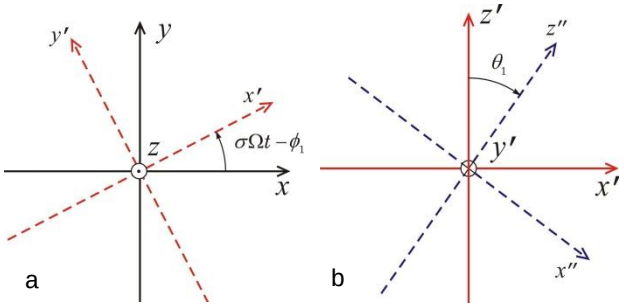

Fig. 2 – The rotating coordinate system: rotation around the *oz* axis (a), followed by rotation around *oy'* (b)

$$A^{-1}=\begin{pmatrix}\cos(\sigma\Omega t-\phi_1) & -\sin(\sigma\Omega t-\phi_1) & 0\\ \sin(\sigma\Omega t-\phi_1) & \cos(\sigma\Omega t-\phi_1) & 0\\ 0 & 0 & 1\end{pmatrix},$$

$$B^{-1}=\begin{pmatrix}\cos\theta_1 & 0 & \sin\theta_1\\ 0 & 1 & 0\\ -\sin\theta_1 & 0 & \cos\theta_1\end{pmatrix}.$$

Then the rotation matrices, according to which the transitions from the rotating system *x''y''z''* to the laboratory one *xyz* are performed, are determined as

$$C^{-1}=A^{-1}B^{-1}=$$
$$=\begin{pmatrix}\cos(\sigma\Omega t-\phi_1)\cos\theta_1 & -\sin(\sigma\Omega t-\phi_1) & \cos(\sigma\Omega t-\phi_1)\sin\theta_1\\ \sin(\sigma\Omega t-\phi_1)\cos\theta_1 & \cos(\sigma\Omega t-\phi_1) & \sin(\sigma\Omega t-\phi_1)\sin\theta_1\\ -\sin\theta_1 & 0 & \cos\theta_1\end{pmatrix}.$$

Consequently, vector $\boldsymbol{\omega}$ and its derivative in the laboratory coordinate system will be written as

$$\boldsymbol{\omega}=C^{-1}\boldsymbol{\omega}''=\begin{pmatrix}-\sigma\Omega\sin\theta_1\cos\theta_1\cos(\sigma\Omega t-\phi_1)\\ -\sigma\Omega\sin\theta_1\cos\theta_1\sin(\sigma\Omega t-\phi_1)\\ \Omega\sin^2\theta_1\end{pmatrix}, \quad (11)$$

$$\dot{\boldsymbol{\omega}}=\begin{pmatrix}\Omega^2\sin\theta_1\cos\theta_1\sin(\sigma\Omega t-\phi_1)\\ -\Omega^2\sin\theta_1\cos\theta_1\cos(\sigma\Omega t-\phi_1)\\ 0\end{pmatrix}. \quad (12)$$

Finally, we represent the vectors M and n, as well as their derivatives, in the system *xyz* taking into account expressions (6) and (7)

$$\mathrm{M}=\begin{pmatrix}M\sin\vartheta_1\cos(\sigma\Omega t-\varphi_1)\\ M\sin\vartheta_1\sin(\sigma\Omega t-\varphi_1)\\ M\cos\vartheta_1\end{pmatrix}, \quad (13)$$

$$\dot{\mathrm{M}}=\begin{pmatrix}-\sigma\Omega M\sin\vartheta_1\sin(\sigma\Omega t-\varphi_1)\\ \sigma\Omega\sin\vartheta_1\cos(\sigma\Omega t-\varphi_1)\\ 0\end{pmatrix}, \quad (14)$$

$$\mathrm{n}=\begin{pmatrix}\sin\theta_1\cos(\sigma\Omega t-\phi_1)\\ \sin\theta_1\sin(\sigma\Omega t-\phi_1)\\ \cos\theta_1\end{pmatrix}, \quad (15)$$

$$\dot{\mathrm{n}}=-\begin{pmatrix}-\sigma\Omega\sin\theta_1\sin(\sigma\Omega t-\phi_1)\\ \sigma\Omega\sin\theta_1\cos(\sigma\Omega t-\phi_1)\\ 0\end{pmatrix}, \quad (16)$$

and introduce the notation

$$F=\mathrm{M}\cdot\mathrm{n}=\sin\theta_1\sin\vartheta_1\cos(\phi_1-\varphi_1)+\cos\vartheta_1\cos\theta_1. \quad (17)$$

We substitute formulas (5), (11)-(17) into (2), equate the expressions in the right and left parts with the same unit vectors. Then, the result obtained for e_x is multiplied by $\cos\sigma\Omega t$, and the result for e_y is multiplied by $\sin\sigma\Omega t$. By summing the found equalities and simplifying the result, we get

$$MV\sin\vartheta_1\sin\varphi_1\left(\sigma\Omega/\gamma-H_{0z}\right)=$$
$$=-\Omega\sin\theta_1\cos\theta_1\left(J\Omega\sin\phi_1+6V\eta\sigma\cos\phi_1\right). \quad (18)$$

Here H_{0z} is the constant field directed along the *oz* axis. Equating terms at e_z, we directly obtain

$$MH\sin\vartheta_1\sin\varphi_1=6\eta\Omega^2\sin^2\theta_1. \quad (19)$$

Similar operations with equation (9) allow to derive

$$MVH_aF\sin(\vartheta_1-\theta_1)\sin\varphi_1=$$
$$=-\Omega\sin\theta_1\cos\theta_1\left(J\Omega\sin\phi_1+6V\eta\sigma\cos\phi_1\right), \quad (20)$$
$$MH_aF\sin\vartheta_1\sin(\varphi_1-\phi_1)=-6\eta\Omega^2\sin\theta_1. \quad (21)$$

In fact, we obtained a system of four equations with respect to unknowns – the angular parameters θ_1, ϕ_1, ϑ_1, φ_1. Let us simplify the derived system by substituting the relation (18) into (20) and the relation (19) into (21). As a result, we write

$$H_aF\sin(\vartheta_1-\theta_1)\sin\varphi_1=\sin\vartheta_1\left(\sigma\Omega/\gamma-H_{0z}\right), \quad (22)$$
$$H_aF\sin\theta_1\sin(\phi_1-\varphi_1)=H\sin\varphi_1. \quad (23)$$

Thus, equations (18), (19), (22), (23) will be considered as the components of the resulting system.

Exact solutions of such a system can be obtained only numerically; however, certain properties of the solutions proceed from the system itself. Thus, it follows from (22) that $\phi_1 > \varphi_1$ and from (23) – that $\vartheta_1 < \theta_1$ for a counterclockwise polarized field ($\sigma = -1$) and no field along the *oz* axis. Then, it follows from (18) that $\theta_1 > \pi/2$ at $\sigma = -1$ and $H_{0z} = 0$. First, this concludes the discussion on the fact that in the precession mode, the angle between the vectors M and H will always be smaller than the angle between n and H. This means that the resulting loss power, which increases with increasing angle between M and H, will be the smaller, the smaller the anisotropy field is. Due to the absence of the magnetic moment motion relative to the crystal lattice, additional losses on account of the magnetic dynamics will be absent; therefore, such a reduction cannot be compensated even theoretically. Second, in the case of finite anisotropy, one can talk about the presence of some (induced by the field of type (5)) effective field along the *oz* axis, equal to $\sigma\Omega/\gamma$, whose direction depends on the polarization of H. When the frequency approaches the ferromagnetic resonance frequency, such an effective field will be comparable to the anisotropy field. This induced field can significantly influence the collective dynamics of an ensemble of ferromagnetic nanoparticles in a rotating field, since it will be a peculiar magnetizing factor, which finally will de-

termine the direction of the resulting dipole field. Therefore, the application of the frozen magnetic moment model can qualitatively distort the hysteresis properties of the nanoparticle ensembles.

At the same time, the questions about the stability of the precessional motion (as in the case of the magnetic moment dynamics in a fixed particle [2]), other possible classes of solutions of equations (1)-(3) and the transitions between them [11] remain open. These questions require a thorough further numerical analysis of the system behavior. Currently, we can say that the preliminary numerical simulation confirms the presence of a stable precession mode, as well as its qualitative characteristics.

4. CONCLUSIONS

Using the equations of motion derived from the total angular momentum conservation law and taking into account the relativity of the magnetic moment motion in the relaxation term [7] to govern the coupled magnetic and mechanical rotational dynamics of a ferromagnetic nanoparticle, we described the precessional motion induced by a circularly polarized external magnetic field of the form (5). The basic procedure of our approach was to find the angular velocity vector in the coordinate system rotating together with the particle, where the given vector has the simplest form, with subsequent transfer to the laboratory coordinate system.

As a result, we obtained the algebraic system of equations for the precession and lag angles of the magnetic moment and the easy axis, which corresponds to the initial equations of motion. The fundamental properties of the solution are the following. 1. The magnetic moment always makes a smaller angle with a rotating field than the anisotropy axis. As a result, there is a decrease in energy losses with decreasing effective anisotropy field. 2. The presence of an effective constant field, equal to $\sigma\Omega/\gamma$ (σ is the factor determining the polarization direction, Ω is the field frequency, γ is the gyromagnetic ratio), perpendicular to the polarization plane, the direction of which is associated with the field rotation direction by the right screw rule. This field in absolute value will tend to the anisotropy field when the frequency approaches the ferromagnetic resonance frequency and can have a significant impact on the collective dynamics of ferromagnetic nanoparticles in a liquid.

The results obtained in the work have theoretical and methodological significance as an important step in the microscopic description of the properties of a ferrofluid. The precession mode can be an initial test example for the subsequent numerical stochastic description of both the single particle dynamics and the collective dynamics of an ensemble of particles. An important applied aspect of the presented results consists in the availability of a more accurate analytical tool for estimating the heating rate of a ferrofluid using an external alternating field in magnetic hyperthermia [10], which is a promising method for treating cancer tumors.

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