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A Stochastic Model for the Dynamics of Superparamagnetic Particles

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Abstract A stochastic model for the dynamics of Superparamagnetic particles based on a classical generalized Lagrangian formalism is proposed. It is a generalization of the model proposed by Brown, allowing for fluctuations on the magnitudes of the magnetic moments of the particles.

1. Introduction

Superparamagnetic particles are very fine particles of a ferromagnetic material, containing a single ferromagnetic domain. Because of their small size,the directions of the magnetic moment show a random time dependence and their magnitudes may also fluctuate around some most probable value. When a magnetic field is applied on a sample of such particles it shows paramagnetic behaviour, with a very big Curie constant, since the individual magnetic moments are several orders of magnitude bigger han the Bohr magneton, μ_B.

The study of the dynamics of this magnetic moment, $\vec{\mu}(t)$, is a very interesting problem in non-equilibrium statistical mechanics, whose conclusions may be verified by suitable experiments, like magnetic resonance[1], Mõssbauer effect[2], etc.

The first stochastic theory proposed for it is due to Brown[3], who postuiated a Langevin type equation obtained from the phenomenological Gilbert's equation[6], by adding to it a noise field term. The weak point of this approach is that it is not suitable for taking into account fluctuations on the magnitudes of $\vec{\mu}$, which may be very important in case of very fine particles. All subsequent theoretical developments derive from Brown's work[3] and suffer from the same drawback.

The basic point to write down an equation of motion for a magnetic moment is to recognize that $\vec{\mu}$ s proportional to some angular momentum and that the time derivative of the angular momentum of any system is equal to the torque applied on it. Well known are the phenomenological equations of Bloch[4], Landau-Lifshitz[5] and Gilbert[6]. We propose now an alternative way of obtaining an equation of motion for $\vec{\mu}(t)$. The entral idea is to assume that the general form of the equation will not depend on the details of the origin of $\vec{\mu}(t)$. Therefore we write $\vec{\mu} = \gamma\vec{S}$, where the angular momentum or "Classical Spin" $\vec{S}$ will be simulated by that of a rotating symmetric body, in the limit of zero moment of inertia and infinite angular velocity, $\dot{\psi}$. By this trick we can write down a classical Lagrangian and derive the equations of motion from it. Landau-Lifshitz equation (or Gilbert's equation) as well as Brown's stochastic equation will follow as articular cases of our general treatment.

Langevin equations

The Lagrangian of a rotating symmetric body is[7]

$$L = \frac{1}{2}I_1\dot{\phi}^2 sen2\theta + \frac{1}{2}I_1\dot{\theta}^2 + \frac{1}{2}I_3(\dot{\psi} + \dot{\phi}cos\theta)^2 - V(\theta,\phi) \tag{1}$$

where I_1 and I_3 are the moments of inertia, 8, ϕ and ψ the usual Euler angles and $V(\theta,\phi)$ some potential energy. The generalized Lagrangian equations of motion read

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{q}_\alpha}\right) - \frac{\partial L}{\partial q_\alpha} = Q_\alpha(t) \tag{2}$$

where the generalized coordinates q_α are in the present case the Euler angles 8, ϕ and ψ, and the "generalized forces" Q_α are the dissipative and stochastic contributions to the total torque, which are not included in the Lagrangian. We assume for Q_α the form[8]

$$Q_\alpha(t) = -\frac{\partial \mathcal{F}}{\partial \dot{q}_\alpha} + N_\alpha(t) \tag{3}$$

where the first term is the dissipative torque, derived from a Rayleigh dissipative function[8]

$$\mathcal{F} = \frac{1}{2}A(\dot{\theta}^2 + \dot{\phi}^2 sin^2\theta) + \mathcal{F}_0(\dot{\psi}) \tag{4}$$

where A is a dissipative constant and $\mathcal{F}_0(\dot{\psi})$ is a function to be defined below. The second term, $N_\alpha(t)$, is the noise torque. When the equations (1), (3) and (4) are substituted into (2) we get a system of three equations of motion. After taking the limit 4 $\rightarrow$ 0, $I_3 \rightarrow 0$ and $\dot{\psi} \rightarrow \infty$ so that $I_3\dot{\psi} = S$ (finite), these equations of motion become

$$A\dot{\theta} + S\dot{\phi}sin\theta = -V_\theta + N_\theta(t) \tag{5.a}$$

$$S\cos\theta - S\dot{\theta}sin\theta + A\dot{\phi}sin^2\theta = -V_\phi + N_\phi(t) \tag{5.b}$$

$$\dot{S} = -B(S - S_0) + N_S(t) \tag{5.c}$$

where $V_\theta = \partial V/\partial\theta$ and $V_\phi = \partial V/\partial\phi$. In the derivation of eq. (5.c) we have assumed that

$$\mathcal{F}_0 = \frac{B}{2}(S - S_0)^2 \tag{6}$$

which may be justified wheever $S(t)$ does not deviate too much from the equilibrium value S_0. The model has, therefore, two relaxation constants, one (A) for relaxation in direction (θ, ϕ) of $\vec{S}(t)$ and the other (B) for relaxation in the magnitude of $\vec{S}(t)$. Correspondingly, we assume also two stochastic torques (noises) $\vec{\xi}(t)$ and $\vec{\Gamma}(t)$, which we will call "transversal noise" and "longitudinal noise", respectively; their cartesian components $\xi_i(t)$ and $\Gamma_j(t)$ will be assumed to behave as independent white noises,

$$< \xi_i(t) > = 0 < \Gamma_j(t) > \tag{7.a}$$

$$< \xi_i(t)\xi_j(t\prime) > = 2D\delta_{ij}\delta(t - t\prime) \tag{7.b}$$

$$< \Gamma_i(t)\Gamma_j(t\prime) > = 2\overline{D}\delta_{ij}\delta(t - t\prime) \tag{7.c}$$

$$< \xi_i(t)\Gamma_j(t\prime) > = 0 \tag{7.d}$$

The spberical noise components, which appear in equations (5), are related to the respective cartesian components by

$$N_\theta = S(\xi_x \cos\theta\cos\phi + \xi_y \cos 0 \sin\phi - \xi_z \sin 0) \tag{8.a}$$

$$N_\phi = S(-\xi_x \sin\theta\sin\phi + \xi_y \sin\theta\cos\phi) \tag{8.b}$$

$$N_S = \Gamma_x \sin\theta\cos\phi + \Gamma_y \sin\theta\sin\phi + \Gamma_z \cos\theta \tag{8.c}$$

By substituting equations (8) into **(5)** we get, after some algebraic manipulation, the following stochastic differential equations for the spherical coordinates (O, ϕ, S) of $\vec{S}$:

$$\dot{q_\alpha} = F_\alpha(q) + \sum_{i=1}^{3} G_{\alpha i}(q)\xi_i(t) + \sum_{i=1}^{3} H_{\alpha i}(q)\Gamma_i(t) \tag{9}$$

where a $= \theta, \phi$, S , i=x, y, z and $F_\alpha G_{\alpha i}$ and $H_{\alpha i}$ are known functions of θ, ϕ and S listed in the Appendix. From the set of equations (9) we can obtain the Fokker-Planck Equation for the probability distribution P(θ, ϕ, S,t), a work which will not be presented here because of space limitation. Some particular cases, however, may be examined easily and are very interesting. Notice, first, that a formal solution to the equation (5.c) is

$$S(t) - S_0 = S(0) - S_0 e^{-\beta t} + e^{-\beta t}\int_0^t e^{\beta t\prime} N_S(t\prime)dt\prime \tag{10}$$

From this equation and the properties

$$< N_S(t) >= 0, < N_S(t)_S(t\prime) >= 2\overline{D}\delta(t - t\prime) \tag{11}$$

which can be easily obtained from equations **(7)** and (8), we get

$$\sigma_S^2(T) =< (\mathrm{S} - S_0)^2 >_{eq} = \lim_{t\to\infty} < (\mathrm{S} - S_0)^2 >= \frac{\overline{\mathrm{D}}}{B} \tag{12}$$

where the equilibrium variance $\sigma_S^2(T)$ is an increasing function of temperature T which depends of the particular features of the particules, like form, volume, crystalline structure, etc, The relation B $= \overline{D}/\sigma_S^2$, for $T > 0$, is the analogous of Einstein relation in the usual Brownian motion, being a manifestation of the Fluctuation-Dissipation Theorem. In the limit of no longitudinal noise, $\overline{D}$ = B = 0, S $= S_0$=constant, the set of equations (5) reduces to two equations for $\theta(t)$ and $\phi(t)$, which are identical to the spherical components of Brown's phenomenological equations[3]

$$\frac{d\vec{\mu}}{dt} = \gamma\vec{\mu} \times \left[\frac{\partial V}{\partial\vec{\mu}} - c\frac{A}{\mu 2}\frac{d\vec{\mu}}{dt} + \vec{h}(t)\right] \tag{13}$$

with the following identifications:

$$h_\theta(t) = \frac{N_\theta(t)}{\gamma S} \tag{14.a}$$

$$h_\phi(t) = \frac{N_\phi(t)}{\gamma S \sin\theta} \tag{14.b}$$

$$h_S(t) = 0 \tag{14.c}$$

Of course, when D $\to$ 0 it follows that we can put $\vec{h}(t) = 0$ and identify equation (14) with the (deterministic) Gilbert equation[6], equivalent to the Landau-Lifshitz (deterministic equation[5]

$$\frac{d\vec{\mu}}{dt} = \gamma_L \vec{\mu} \times \vec{H} - \frac{\lambda_L}{\mu^2}\vec{\mu} \times (\vec{\mu} \times \vec{H}) \tag{15}$$

where $\vec{H} = -\,\partial V/\partial\vec{\mu}$, $\vec{\mu} = \gamma\vec{S}$, $\gamma_L = \frac{\gamma S^2}{A^2+S^2}$ and $\lambda L = \frac{AS\gamma^2}{A^2+S^2}$.

In conclusion we may say tbat, by a procedure based on classical analytical mechanics, we obtained general equations for the dynamics of the magnetic moments of single domain particles, which extend previous results, allowing for more general fluctuations, and reduce to them when the appropriate contraints are imposed.

Appendix A

$$F_\theta = -a(S)V_\theta + \frac{b(S)}{\sin\theta}V_\phi - Bb(S)(S - S_0)ctg\theta$$

$$F_\phi = -\frac{b(S)}{\sin\theta}V_\theta - \frac{aS)}{\sin^2\theta}V_\phi - Ba(S)(S - S_0)\frac{\mathrm{ctge}}{\mathrm{sin}\ 19}$$

$$F_S = -B(S - S_0)$$

$$a(S) = \frac{A}{A^2 + S^2}$$

$$b(S) = \frac{S}{A^2 + S^2}$$

$$G_{\theta x} = Sa(S)\cos\theta\cos\phi + b(S)\sin 4$$

$$G_{\theta y} = Sa(S)\cos\theta\sin\phi - b(S)\cos\phi$$

$$G_{\theta z} = -Sa(S)\sin\theta$$

$$G_{\phi x} = S\frac{b(S)\cos\theta\cos 4 - a(S)\,\text{sín}\,\phi}{\sin\theta}$$

$$G_{\phi y} = S\frac{b(S)\cos\theta\sin 4 + a(S)\cos\phi}{\sin\theta}$$

$$G_{\phi z} = -Sb(S)$$

$$G_{Sx} = G_{Sy} = G_{Sz} = 0$$

$$H_{\theta x} = Sb(S)\cos\theta\cos\phi$$

$$H_{\theta y} = Sb(S)\cos\theta\sin\phi$$

$$H_{\theta z} = Sb(S)ctg^2\theta\sin\theta$$

$$H_{\phi x} = -Sa(S)ctg\theta\cos\phi$$

$$H_{\phi y} = -Sa(S)ctg\theta\sin\phi$$

$$H_{\phi z} = -Sa(s)ctg^2\theta$$

$$H_{Sx} = \sin\theta\cos\phi$$

$$H_{Sy} = \sin\theta\sin\phi$$

$$H_{Sz} = \cos\theta$$

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Resumo

Propomos um modelo estocástico para a dinâmica de partículas Superparamagnéticas baseado em formalismo Lagrangeano clássico generalizado. Trata-se de uma generalização do modelo proposto por Brown, que permite flutuações nas magnitudes dos momentos magnéticos das partículas.