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Superparamagnetic relaxation time under the effect of a randomly oriented magnetic field for weakly interacting ultrafine particles

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Abstract

The effect of a constant and a randomly oriented magnetic field H(ψ,ϕ), on the Néel relaxation time τ, of an assembly of weakly interacting ultrafine ferromagnetic particles with uniaxial anisotropy, is investigated. In this regard, the resolution of the Fokker-Panck equation (FPE) for the probability density W(θ,φ,t) of an assembly of ultrafine ferromagnetic particles, with uniaxial anisotropy in the presence of an applied field H(ψ,ϕ) was performed and published as a first part of this work; the results generalize previous calculations, and give satisfactory interpretation of the relaxation phenomenon in such system. In this part, calculation of the relaxation time, τ, is performed under the assumption of weak interactions between particles. The derived expression of τ is in good concordance with those previously obtained in the first part, and is consistent with those of Coeffey et al. for high values of the anisotropy parameter.

1. Introduction

Superparamagnetique ultrafine particles have received much attention since several decades due to the wide possibilities of their technological applications, and to fundamental questions on the basic of some observed phenomena [1-9].

The effect of an external constant magnetic field, H, on the longitudinal relaxation time, τ, of the ferromagnetic nanoparticales having simple uniaxial anisotropy may be studied by calculating the smallest nonvanishing eigenvalue λ_1 (the escape rate) of the Fokker-Planck equation (FPE).

Relaxation time of ultrafine particles with uniaxial symmetry has been evaluated, from Brown's model, by Aharoni and Eisenstien [10, 11], Jones and Srivastava [12] and Bessais et al. [13], with the hypothesis that the external magnetic field is zero or parallel to the magnetization easy direction axis (e_3). Later, these calculations were done by Coffey et al. and Mørup et al. in the case where the external magnetic field was supposed to be oblique H(ψ) [14-16]. Recently, we have extended calculations to the case of an aleatory magnetic field H(ψ,ϕ) [17].

In this work, we used the mathematical method of discretisation to solve the asymptotic analytical formula of the superparamagnetic relaxation time under the effect of a randomly oriented magnetic field for weakly interacting ultrafine particles.

2. Mathematical model and method

2.1. Brown's model

Fokker-Planck equation governs the time evolution of the density of magnetic moment orientation W(θ,φ,t) on a sphere of radius Ms (the mean magnetization of a nonrelaxing particle), the orientation of the magnetic moment

M being specified by the spherical polar coordinates (θ,φ). For simple uniaxial anisotropy the ratio of the potential energy vV to the thermal energy K_BT may be expressed by the following equation:

$$\beta V = \alpha\left[1-(re_3)^2\right] - \xi\,(re_H) \tag{1}$$

In this equation, $\beta=v/K_BT$, v denoting the volume of the nanoparticule; $\alpha=Kv/K_BT$ represents the anisotropy parameter, K_BT is the thermal energy; K the anisotropy constant, $\xi=vM_SH/K_BT$ is the external field parameter, r, e_3 and e_H are unit vectors in the direction of the magnetization vector M, the internal anisotropy (or easy) axis, and H respectively. $e_H = H/\|H\| = \sin\psi\cos\phi e_1 - \sin\psi\sin\phi e_2 + \cos\psi e_3$

The Néel equation time, which is the time required for the magnetization to surmount the potential barrier given by Eq (1), is related to λ_1 by the following equation [18, 19]:

$$\tau \cong \frac{\tau_N}{\lambda_1} \tag{2}$$

where λ_1 is the smallest non-vanishing eigenvalue of the FPE and τ_N is the diffusional relaxation time given by [19].

$$\tau_N = \frac{\beta\eta M_s^2\left(1+a^2\right)}{2a^2} \tag{3}$$

In this equation a=ηγMs, with units such that ''a'' is a dimensionless damping parameter, in which η is the phenomenological damping constant from Gilbert's equation [20], and γ denotes the gyromagnetic ratio.

2.2. Numerical calculation of the smallest non-vanishing eigenvalue λ_1 of the FPE

The probability density distribution, W(θ,φ,t), of an assembly of ultrafine ferromagnetic particles of volume v, with uniaxial anisotropy in the presence of an applied field H(ψ,ϕ) satisfies the FPE, and is given, in spherical coordinates, by [19] :

$$\dot{W} = L_{FP} W \tag{4}$$

where

$$L_{FP} W = \beta^{-1} b\Delta W + bW\Delta V + b\left(\frac{\partial V}{\partial\theta}\frac{\partial W}{\partial\theta} + \frac{1}{\sin^2\theta}\frac{\partial V}{\partial\varphi}\frac{\partial W}{\partial\varphi}\right) + \frac{b}{a\sin\theta}\left(\frac{\partial V}{\partial\theta}\frac{\partial W}{\partial\varphi} - \frac{\partial V}{\partial\varphi}\frac{\partial W}{\partial\theta}\right) \tag{5}$$

In this equation Δ is the Laplacian in spherical coordinates and $b = a\,\gamma/(1+a^2)Ms$.

In the case of uniaxial symmetry, the anisotropy energy (Eq (1)) may be written as :

$$\beta V = \alpha\sin^2\theta - \xi\cos\theta\cos\psi - \xi\sin\theta\cos\varphi\sin\psi\cos\phi + \xi\sin\theta\sin\varphi\sin\psi\sin\phi \tag{6}$$

The potential of Eq. (6) is non-axially symmetric unless if ψ=0, so the gyroscopic terms expressed in (a^{-1}) in Eq. (5) will not vanish since (a) is of the order of 0.2 to 1. We will ignore these terms in a first approximation [15]. Eq. (4) takes then the following form :

$$2\,\tau_N\frac{dW}{dt} = \Delta W + W\Delta\,(\beta V) + \left(\frac{\partial(\beta V)}{\partial\theta}\frac{\partial W}{\partial\theta} + \frac{1}{\sin^2\theta}\frac{\partial(\beta V)}{\partial\varphi}\frac{\partial W}{\partial\varphi}\right) \tag{7}$$

where $\tau_N=\beta/2b$ denotes the diffusional relaxation time.

λ_1 is calculated by expanding the solution of Eq (7) as a series of spherical harmonics whence the calculation reduces to a matrix eigenvalue problem. This is described in detail in Ref [17].

3. Asymptotic expression for λ_1

Consider a monodomain particle (i) of uniaxial symmetry, with magnetic moment μ_i . The other particles of the system act on (i) via a dipolar field B_i oriented at an angle ψ with e_3 considered as the easy magnetization axis (figure 1) :

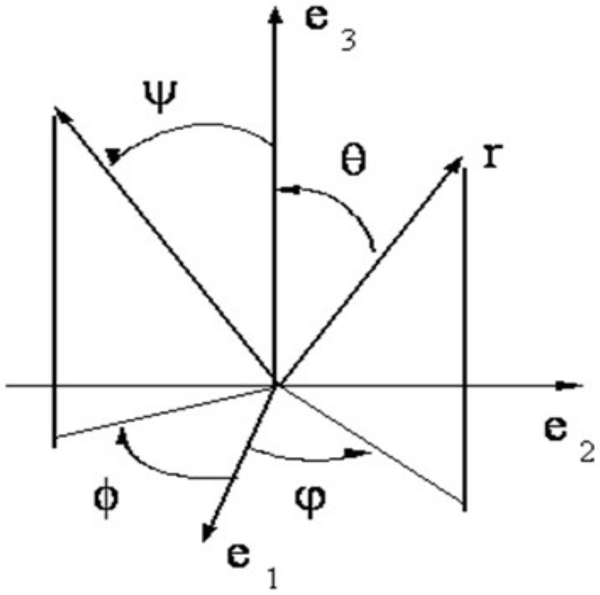

Figure 1 : The field and magnetization orientations in terms of spherical polar coordinates.

The anisotropy energy can be expressed as:

$$E = K v \sin^2\theta - \vec{\mu}_i \vec{B}_i \tag{8}$$

With the following variables change $\xi = \mu_i B_i/K_B T$, this energy may be written in the following form:

$$\beta E = \alpha \sin^2\theta - \xi\left[\cos\theta\cos\psi + \sin\theta\cos\varphi\sin\psi\cos\phi - \sin\theta\sin\varphi\sin\psi\sin\phi\right] \tag{9}$$

We can express the global minimum energy and the local maximum energy in the limit of weak interactions in which $\xi <<1$. In this limit, minimum and maximum values are obtained for $\theta \approx 0$ and 90 respectively.

When θ is near to zero, sin θ ≈ tg θ

Expressing the derivative of Eq. 9 with respect to θ, $(\partial\beta E/\partial\theta)=0$, gives:

$\sin\theta \cong h\,[(\sin\psi\cos\phi)/(1+h\cos\psi)]$, where $h=\xi/2\alpha$. $h<<1$, so $\sin\theta \approx h \sin\psi\cos\phi$,

Consequently the minimum value of energy when θ is near to zero, is :

$$\beta E_{min} = -\alpha\left[h^2\sin^2\psi\cos^2\phi + 2h\cos\psi\right] \tag{10}$$

When θ is near to 90°, a similar approximation to that used above leads to $\cos\theta \approx -h\cos\psi$, and then:

$$\beta E_{max} = \alpha\left[1 + h^2\cos^2\psi - 2h\cos\varphi\sin\psi\cos\phi + 2h\sin\varphi\sin\psi\sin\phi\right] \tag{11}$$

The energy barrier $\Delta E = E_{max} - E_{min}$ can be written as:

$$\beta\Delta E = \alpha\left[1 + h^2\left(1 - \sin^2\psi\sin^2\phi\right) + 2h\cos\psi - 2h\cos\varphi\sin\psi\cos\phi + 2h\sin\varphi\sin\psi\sin\phi\right] \tag{12}$$

The transition probability to overcome the energy barrier is given by :

$$f(\psi,\varphi,\phi) = (2\pi\tau_0)^{-1}\exp(-\beta\Delta E) \tag{13}$$

Integration of f (ψ,ϕ,φ) with respect to φ, gives:

$$f(\psi,\phi) = \tau_0^{-1}\exp-\alpha\left[1 + h^2\left(1 - \sin^2\psi\sin^2\phi\right)\right] \times \left\{1 - \xi\cos\psi + \left(\frac{\xi}{2}\right)^2\left(1 + \cos^2\psi\right)\right\} \tag{14}$$

And the relaxation time is given by:

$$\tau^{-1} = \frac{\iint f(\psi,\phi)\sin\psi\, d\psi\, d\phi}{\iint \sin\psi\, d\psi\, d\varphi} \tag{15}$$

An integration first with respect to ψ gives:

$$\tau^{-1} = \frac{\tau_0^{-1}}{4\pi}\int_0^{2\pi} \exp-\alpha\left(1+h^2\cos^2\phi\right) \times \left[2+\left(\frac{\langle\xi\rangle}{2}\right)^2\left(\frac{8}{3}-\frac{2}{3}\alpha h^2\sin^2\phi\right)\right]d\phi \quad (16)$$

and then with respect to ϕ leads to:

$$\tau = \frac{\tau_0}{I_0\left(\alpha h^2/2\right)}\exp\alpha\left[1-\frac{4}{3}\langle h^2\rangle\left(\alpha-\frac{1}{2}\right)\right] \quad (17)$$

where I_0 is the Bessel's function such as: $I_0(x)(x \ll 1) = 1+\frac{(x/2)^2}{(1!)^2}+\frac{(x/2)^4}{(2!)^2}+....$

Finally τ can be expressed as:

$$\tau = \frac{\tau_0}{1+\left(\frac{\alpha\langle h^2\rangle}{4}\right)^2}\exp\left[\alpha-\frac{\langle\xi^2\rangle}{3}\left(1-\frac{1}{2}\alpha^{-1}\right)\right] \quad (18)$$

Where the parameter τ_0 may be considered as approximately independent of the magnetic field [13]:

$$\tau_0 = \frac{1}{2}\beta b^{-1}\left(1+\frac{\alpha}{4}\right)^{-5/2} \quad (19)$$

The smallest nonvamling eigenvalue of the FPE may be expressed as a function of the relaxation time :

$$\lambda_1 \cong \tau_N\tau^{-1} = \frac{\beta b^{-1}\tau^{-1}}{2} \quad (20)$$

The asymptotic expression λ_A, of λ_1, may be derived from Eq (20) with the help of Eqs (19) and (3), such as:

$$\lambda_A = \left(1-\frac{\alpha h^2}{6}+\frac{4\alpha^2h^2}{3}\right)\left(1+\frac{\alpha}{4}\right)^{5/2}\exp-\alpha\left[1-h^2\left(\frac{4}{3}\alpha-1\right)\right] \quad (21)$$

$$\lambda_A = \left(1-\frac{\alpha h^2}{6}+\frac{4\alpha^2h^2}{3}\right)\left(1+\frac{\alpha}{4}\right)^{5/2}\exp-\alpha\left(1+h^2\right) \quad (22)$$

Eigenvalues were also calculated using the Linalg instruction and Maple library. The smallest non vanishing eigenvalue was determined using the min (c, X [j]) instruction. The matrix order was 230*230.

Figures 2, give a comparison between the variation, as a function of α and for different values of h, of the asymptotic expression, λ_A, of λ_1 (Eq.(22)) and the numerical simulation. The corresponding curves show a very good concordance in the case of weak fields.

Symptotic formula proposed by Coffey et al. is given by [16] :

$$\lambda_A = \frac{\alpha(1-h)(1+h)^{1/2}}{\pi h}\exp-\alpha(1-h)^2 \quad (23)$$

Our asymptotic formula matches with that of Coffey et al. for α>5 as shown on figures 3 :

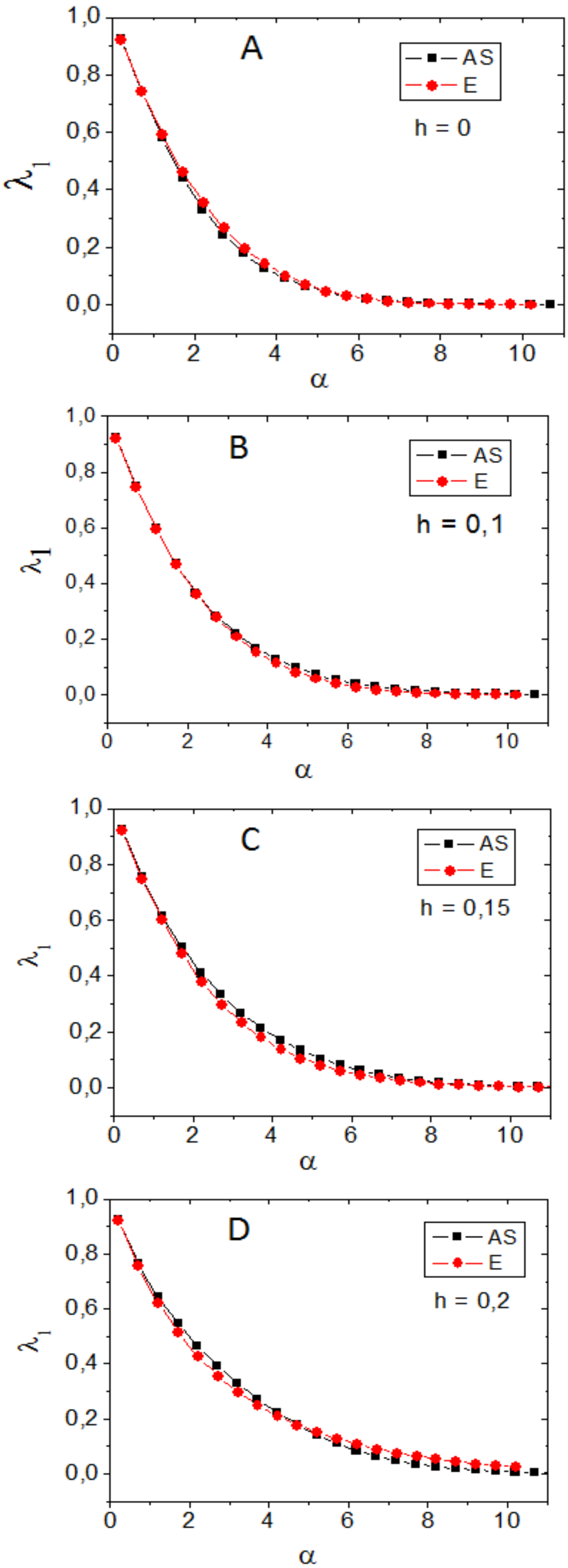

Figure 2: *Variation of λ_1 as a function of α, for ψ= 90 and ϕ=90, and for:* ***A****- h=0,* ***B****- h=0.1,* ***C****- h=0.15, D- h=0.2*
AS*: asymptotic formula,* ***E****: numerical simulation*

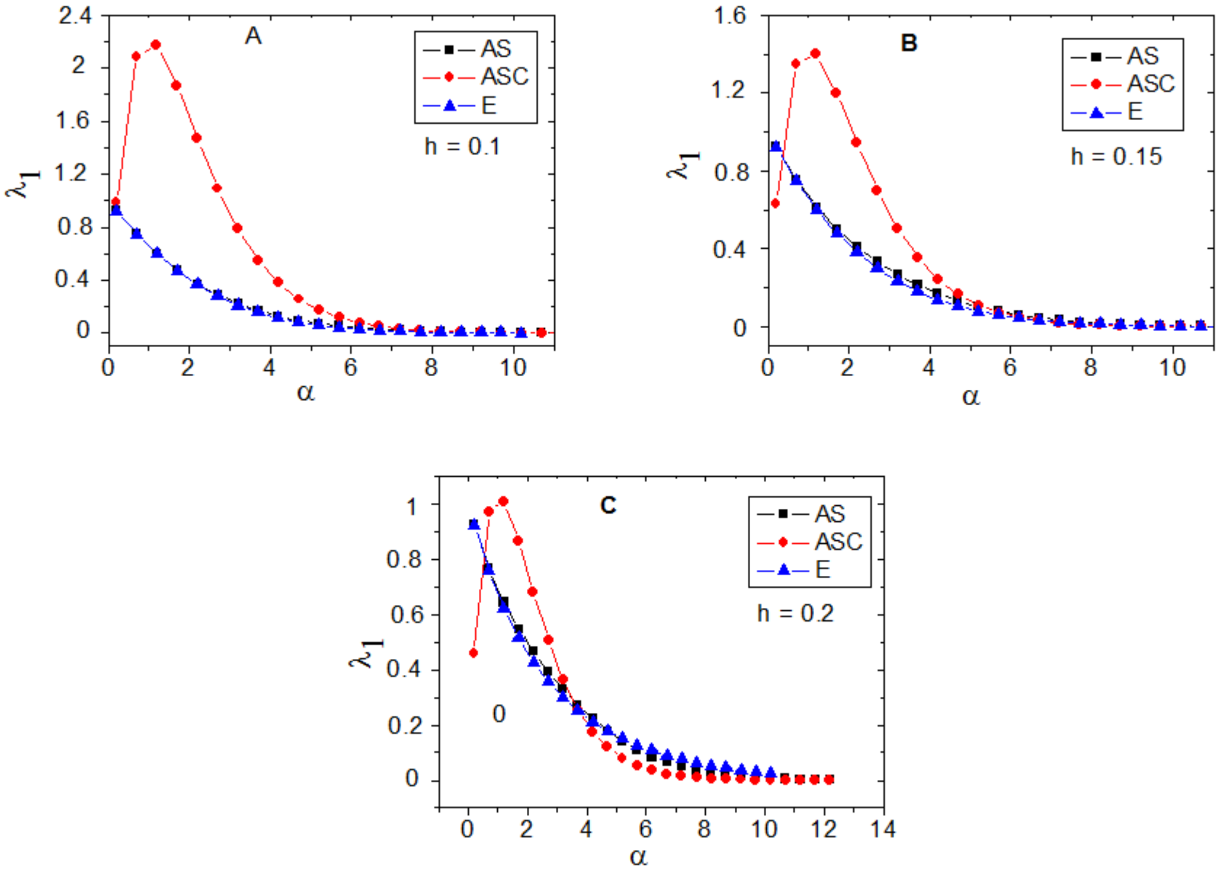

Figure 3: *Variation of λ_1 as a function of α, for ψ= 90 and ϕ=90, and for:* ***A****- h=0.1;* ***B****- h=0.15;* ***C****- h=0.2.* ***AS****: asymptotic formula,* ***E****: numerical simulation,* ***ASC****: asymptotic formula (Coffey et al.)*

Conclusion

An asymptotic analytical formula of the relaxation time of an assembly of ultrafine particles with uniaxial anisotropy experiencing a random oriented magnetic field has been derived. Results obtained are in good concordance with those previously derived from a numerical approach, and published elsewhere [17]. Moreover, our results disagree with those of Coffey et al. for anisotropy parameter (α) values inferior approximately to 6.

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