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Stochastic resonance in single-domain particles

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Abstract. A simple approximate equation of motion for the longitudinal component of the magnetic moment of a single-domain uniaxial superparamagnetic particle is proposed. This equation is used to evaluate the spectral density function of the interparticle magnetic motion at finite temperatures and to describe the stochastic resonance effect. The correspondence of the results obtained with those of previous calculations is discussed.

1. Introduction

The phenomenon of stochastic resonance (SR) predicted not long ago [1] in the noise-driven behaviour of multi-stable systems, has since attracted a considerable attention [2–5]. As is always the case with effects related to Brownian motion, SR has a wide range of applicability spreading from geometeorology [1] to laser physics [2].

The manifestation of SR is rather simple. To a bistable system, subjected to noise, a weak alternating field (modulation) of a frequency Ω favouring the transitions between the equilibria is applied. Under these conditions the signal-to-noise ratio, determined from the spectral density function $Q(\omega)$ at $\omega = \Omega$ as a function of temperature, passes through a distinctive maximum. One of the peculiar features of SR is that its existence does not depend upon the actual type of dynamic equation as long as modulation is additive. In other words, SR is not sensitive to whether the character of motion is oscillatory or relaxational.

In recent papers [6, 7] the SR theory was applied to a very clear physical situation: a uniaxially anisotropic single-domain ferromagnetic particle. In the absence of interaction with the neighbours its magnetic energy is

$$U = -\mu H\,(\boldsymbol{e}\boldsymbol{h}) - KV\,(\boldsymbol{e}\boldsymbol{n})^2 \tag{1}$$

where $\boldsymbol{e}$, $\boldsymbol{n}$ and $\boldsymbol{h}$ are the unit vectors of the particle magnetic moment, the anisotropy axis and the external field, respectively; K is the effective anisotropy constant (for uniaxial anisotropy it is essentially positive), $\mu = I_s V$ is the magnetic moment of a single-domain particle, I_s is its magnetization and V its volume. As it apparent from (1), without external fields the component of the magnetic moment $\mu(\boldsymbol{e}\boldsymbol{n})$ along the anisotropy axis has two, perfectly equal in energy, equilibrium orientations, viz., $\boldsymbol{e} \parallel \boldsymbol{n}$ and $\boldsymbol{e} \parallel -\boldsymbol{n}$, thus making a one-dimensional bistable system. The rate of transition between those potential wells is controlled by the parameter [8]

$$\sigma = KV/k_B T$$

which, assuming that the barrier height KV is fixed, one may regard as the dimensionless inverse temperature.

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In the present paper we revise and extend the results of [6, 7]. Namely, hereby we

—propose a simple equation of motion for the longitudinal component of the magnetic moment of a superparamagnetic particle, where the relaxation time is a function of the potential barrier,

—demonstrate that in a wide temperature range this relaxation time with high accuracy *coincides with that of the lowest mode of the pertinent Fokker–Planck equation,*

—evaluate SR using the adopted equation for the longitudinal relaxation, and compare the resulting curve with those based on recently proposed simple approximate expressions for the superparamagnetic relaxation time,

—show that the description derived from the kinetic equation approach enables one to evaluate SR eliminating the quasi-static and high-barrier approximations on which previous calculations were essentially based and

—explain that the method of two alternating fields proposed in [6, 7] may not be used for direct observation of SR.

2. Equation of motion for the longitudinal component of the magnetic moment

The linear magnetodynamics of a single-domain superparamagnetic particle was studied in [9, 10], where a Fokker–Planck-like rotary diffusion equation for the orientational distribution function $W(e, t)$ of the unit vector $e = \mu/\mu$ of the magnetic moment was derived. Though the initial Langevin equations of the problem were slightly different—based on the Gilbert equation in [9] and on the Landau–Lifshitz one in [10]—with the accuracy of notations the results concerning the longitudinal component of e coincide. At $\sigma \neq 0$ (anisotropic particle) the free motion of the observable, i.e., averaged over the statistical ensemble, magnetic moment $\langle e \rangle n$ is described by a discrete infinite spectrum of relaxation times.

With the aid of the dimensionless decrements $\lambda_l(\sigma)$ of the corresponding boundary problem [10] the set of those relaxation times may be presented as

$$\tau_l(\sigma) = 2\sigma\tau_0/\lambda_l(\sigma) \tag{2}$$

where the subscript l enumerates the eigenmodes of the orientational distribution function connected with the longitudinal (with respect to n) motions. The temperature-independent characteristic time $\tau_0 = I_s/2\alpha\gamma K$ determines the decay rate of the Larmor precession of the magnetic moment in the uniaxial anisotropy field $H_a = 2K/I_s$ of the particle at low temperature. In the equivalent form it may be rewritten as $\tau_0 = (\alpha\omega_0)^{-1}$, where $\omega_0 = \gamma H_a$ is the Larmor frequency (γ being the gyromagnetic ratio) and α is the dimensionless phenomenological parameter [11] describing precession damping in the framework of the Landau–Lifshitz equation.

The exact spectrum $\{\lambda_l(\sigma)\}$ can be evaluated only by an elaborate numerical procedure involving a large number of eigenmodes [10], but the asymptotic expressions are simple [9, 10]:

$$\lambda_l(\sigma \ll 1) = l(l+1) + O(\sigma) \qquad \lambda_l(\sigma \gg 1) \propto \begin{cases} \sigma^{3/2}e^{-\sigma} & \text{for } l = 1 \\ 2\sigma & \text{for } l = 2, 3, \ldots \end{cases} \tag{3}$$

Formulae (3) are sufficient to reveal the qualitative difference between λ_1 and all the other decrements. Obeying the common expression $\lambda_l = l(l+1)$ at $\sigma \to 0$, in the opposite limit

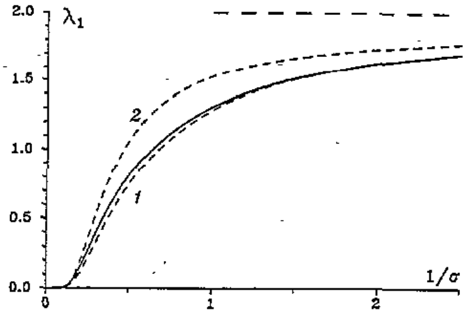

Figure 1. The first eigenvalue as a function of the dimensionless temperature parameter $1/\sigma = k_B T/KV$. Curve *1*—numerical evaluation, 2—by interpolation formula (15) after [12],, *3*—by interpolation formula (16) after [13].

λ_1 is exclusive—it exponentially goes down whereas all the others display linear growth. The exact behaviour of $\lambda_1(\sigma)$ at intermediate σ is shown in figure 1 by a solid line. Two dashed lines there resemble the approximate expressions for λ_1 recently proposed by Bessais *et al* [12] and Aharoni [13].

The relaxation time associated with λ_1 is singled out as well. According to equation (2), it becomes exponentially large

$$\tau_1 \sim \tau_0\, e^{\sigma}$$

at high σ thus signalling of slowing down (blocking) of the particle magnetic moment transitions between n and $-n$ at low temperatures. However, to know to what extent τ_1 really affects the motion of the particle magnetic moment, we have to evaluate the weight coefficients with which different modes of $W(e, t)$ enter the exact expression for $\langle e(t)\rangle n$.

The corresponding calculation, which is highlighted in the appendix, shows that for the longitudinal projection of $\langle e\rangle$ the contribution of the lowest, i.e., with $l = 1$, mode is absolutely dominating. This result, not at all obvious until having been proven, justifies the effective closure of the infinite set of equations for the moments $\langle e_i\rangle$, $\langle e_i e_k\rangle$, ... of the distribution function W (see equation (A2) of the appendix) and leads to a single self-sufficient equation for the projection of $\langle e\rangle$ on n:

$$\tau_1 \frac{d}{dt} m + m = \frac{\mu^2 B}{k_B T} H \tag{4}$$

where $m = \mu\langle e\rangle n$ and the temperature-dependent coefficient $B = d\ln Z/d\sigma$ is defined via the integral $Z(\sigma)$—see formula (A4) of the appendix. The numerical value of B grows from $\frac{1}{3} + O(\sigma)$ at $\sigma \ll 1$ (low potential barrier) up to $1 - O(\sigma^{-1})$ at $\sigma \gg 1$ (high potential barrier). We remark that the equation obtained is already linearized with respect to the applied external field H. The details of derivation of the right-hand side of equation (4) may be found in [10].

3. Spectral density function

For the cases where only the longitudinal component of the magnetic moment is relevant, equation (4) provides a very simple and accurate reduction of the complete rotary diffusion equation. In principle, a similar approximating procedure may be carried out to obtain a simplified representation for the time-correlation function $\mu^2\langle e_i(t)\, e_k(t')\rangle n_i\, n_k$ of the longitudinal projection. The pertinent initial condition for this function readily follows from the equilibrium distribution $\exp(-U/k_\mathrm{B}T)$ at $H = 0$. In the notations of equation (4) it reads

$$\langle m^2(t)\rangle = \mu^2 B\,. \tag{5}$$

However, those cumbersome calculations may be circumvented using the method of the Langevin equation (see [14]) introducing into equation (4) an effective white noise. This modification gives

$$\tau_1 \frac{\mathrm{d}}{\mathrm{d}t}\, m + m = \frac{\mu^2 B}{k_\mathrm{B} T}\,(H + F) \tag{6}$$

where for the auxiliary random magnetic field with allowance for equation (5) one obtains

$$\langle F(t) F(t')\rangle = 2D\,\delta(t - t') \qquad 2D = 2\tau_1 k_\mathrm{B}^2 T^2/\mu^2 B\,. \tag{7}$$

Now applying to the system the oscillating magnetic field of frequency Ω, i.e., setting in equation (6)

$$H(t) = \tfrac{1}{2} H\, \left[\mathrm{e}^{\mathrm{i}(\Omega t+\varphi)} + \mathrm{e}^{-\mathrm{i}(\Omega t+\varphi)}\right] \tag{8}$$

where φ is an arbitrary initial phase of the oscillation, and making the time-frequency Fourier transformation, we obtain

$$m_\omega = \frac{\mu^2 B}{k_\mathrm{B} T}\, \frac{1}{1 - \mathrm{i}\omega\tau_1}\, \left\{F_\omega + \pi\, H\, \left[\mathrm{e}^{\mathrm{i}\varphi}\,\delta(\omega + \Omega) + \mathrm{e}^{-\mathrm{i}\varphi}\,\delta(\omega - \Omega)\right]\right\}. \tag{9}$$

With the use of the relation

$$\langle x_\omega x^*_{\omega'}\rangle = 2\pi\, \left(x^2\right)_\omega \delta(\omega - \omega')$$

connecting the product of the Fourier transforms with the corresponding spectral density, taking into account that the noise-induced field F is random and performing the averaging over φ, we transform equations (7), (9) into the expression

$$\left(m^2\right)_\omega = \left(\frac{\mu^2 B}{k_\mathrm{B} T}\right)^2 \left\{\frac{2D}{1 + \omega^2\tau_1^2} + \frac{\pi\, H^2}{2(1 + \Omega^2\tau_1^2)}\, [\delta(\omega - \Omega) + \delta(\omega + \Omega)]\right\} \tag{10}$$

rendering the spectral density of motion of the component of magnetic moment along the anisotropy axis. Inside the curly brackets on the right-hand side of equation (10) the first term should be identified with the thermal noise. It describes the energy spectrum of spontaneous orientational transitions between the potential wells created by the magnetic anisotropy. The second term, which is formally singular, is proportional to the power of the applied field and thus yields the contribution of the induced signal.

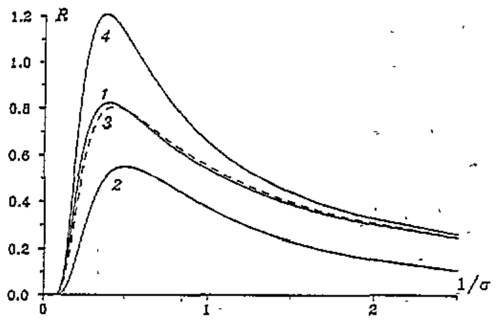

Figure 2. Dimensionless SR curves, i.e., functions $R(\sigma)$ of equation (13). Line 1—based on the numerical calculation of λ_1; line 2—from equation (14); line 3 (dashed)—with λ_1 by equation (15); line 4—with λ_1 by equation (16).

4. Stochastic resonance

Let us consider the spectral density function $Q(\omega) \equiv (m^2)_\omega$ at the frequency of the external field, i.e., at $\omega = \Omega$. Replacing, as in [3], the symmetrized Fourier transforms by the one-sided ones, from equation (10) we obtain

$$\frac{k_B T}{\mu^2 B} Q(\Omega) = \frac{2}{1+\Omega^2\tau_1^2} \left[2D + \tfrac{1}{2}\pi H^2 \delta(\omega - \Omega)\right]. \tag{11}$$

According to the conventional definition of the stochastic resonance [3, 4], it appears when, splitting the spectral density into two qualitatively different parts, one, constructs the signal-to-noise ratio. For the function (11) the choice is obvious, and thus the signal-to-noise factor is

$$S = \pi H^2/4D = \pi H^2 \mu^2 B / 4\tau_1 k_B^2 T^2 \tag{12}$$

here we have made use of formula (7).

To study the temperature dependence of the SR characteristic S, it is convenient to rewrite the latter once more, grouping the temperature-sensitive functions. With formulae (2) and $\mu = I_s V$ we arrive at the representation

$$S = \frac{\pi}{8\tau_0}\left(\frac{I_s H}{K}\right)^2 R \qquad R(\sigma) \equiv \sigma B(\sigma)\lambda_1(\sigma) \tag{13}$$

which shows that the subject of our principal interest is the dimensionless function R of the dimensionless argument $\sigma = KV/k_B T$. The presence of SR follows immediately from its asymptotics. Using equations (3) and the limiting expressions for $B(\sigma)$ one finds

$$R(\sigma) = \begin{cases} \frac{2}{3}\sigma & \text{for } \sigma \to 0 \\ \sigma^{5/2} e^{-\sigma} & \text{for } \sigma \to \infty \end{cases}$$

which indicates that R assumes its maximum at finite σ. The qualitative explanation is apparent: at very high temperatures ($\sigma \to 0$) the thermal noise takes over any regular motion, whereas at very low temperatures ($\sigma \to \infty$) the potential wells tightly trap the magnetic moments.

The actual form of the SR curve obtained with the aid of formula (13) is shown in figure 2; the location of the maximum is $\sigma \simeq 2.5$. For comparison we have also plotted there the SR curve of [6]. (Though the contents of the [6,7] coincide almost exactly, their resulting SR functions differ. It seems that the latter paper contain some misprints. For example, the expression for the transition rates there is obviously wrong.) Rewritten in the notations adopted here, the characteristic function R of [6] reads

$$R(\sigma) = \left(2^{5/2}/\pi^{3/2}\right) \sigma^2 \exp(-\sigma) \tag{14}$$

and predicts the maximum at $\sigma = 2$. Note that while deriving equation (13) we have used neither the adiabaticity assumption $\omega\tau_0 \ll 1$ nor the high-barrier approximation $\sigma \gg 1$. Both these conditions were essentially used in [6,7] while deriving to formula (14). However, the comparison shows that disagreement between the exact and approximate results is not too dramatic, since the locations of the maxima differ by not more than 20%. Two other curves (*3* and *4*) in figure 2 display the results of using, instead of the exact numerical values of $\lambda_1(\sigma)$, the interpolation expressions, namely,

$$\lambda_1(\sigma) = 2(1+\sigma/4)^{5/2} \exp(-\sigma) \tag{15}$$

proposed in [12], and

$$\lambda_1(\sigma) = 2\left(\frac{2+9\sigma/5+(4/\pi)^{1/3}\sigma^2}{2+\sigma}\right)^{3/2} \exp(-\sigma) \tag{16}$$

proposed in [13]. As in figure 1, approximation (15) provides much better results than the other one.

As to the observation and measurement of the SR curve, the direct way to them is pointed out by expressions (10)–(11). The signal-to-noise ratio may be obtained either directly by evaluating $Q(\omega)$ from the autocorrelation function $\langle m(t)\, m(0)\rangle$ or calculated from the data on complex susceptibility [15].

In [6,7] another method to obtain the SR data has been proposed. It is claimed that if, besides $H(t)$, one imposes on the system one more additional alternating field (in [6,7] it is called the 'probing field')

$$H_1(t) = \tfrac{1}{2} H_1 \left[e^{i(\omega_p t+\psi)} + e^{-i(\omega_p t+\psi)} \right]$$

—cf. equation (8)—also weak and parallel to the anisotropy axis, and measure the imaginary part of the corresponding susceptibility $\chi(\omega_p, \Omega) = \partial m/\partial H_1$, then the imaginary part of the dynamic response $\chi(\omega_p, \Omega)$ to the field H_1 is proportional to the complete spectral density of the system subjected to the magnetic field $H(t)$. It therefore contains all the sufficient information to obtain the SR curve. From the viewpoint of the author of [6,7], the proportionality $\chi(\omega_p, \Omega) \propto Q(\omega_p)$ at $H \neq 0$ follows from the fluctuation-dissipation theorem (FDT).

However, this is not so. To understand this, one has just to recall that FDT holds only for the noise component of the spectral density function. That means that $\Im\chi(\omega_p, \Omega)$ obtained by the method solicited in [6,7] will yield merely the part of $Q(\omega_p)$, proportional to temperature, i.e., noise, as if the field $H(t)$ had not existed at all. In other words, involvement of one more oscillating field is unable to provide any additional information. In the correct approach, the solution of the problem is simple. For evaluation of the SR curve, it is sufficient to have the data on the usual dynamic susceptibility with respect to a single probing field, no matter whether H or H_1. As is shown in [15], the signal-to-noise ratio may be exactly expressed in terms of real and imaginary parts of this function.

5. Conclusion

We have shown that for a physically important case of uniaxially anisotropic superparamagnetic particles convenient for observation the motion of the longitudinal projection of the magnetic moment may be rather accurately described with a single closed equation of relaxational type. The characteristic time entering this equation is defined for the whole temperature range, at low temperatures being very close to the conventional one [9] determining the superparamagnetic relaxation in the $\cos^2\vartheta$ orientational double-well potential at considerable heights of the barrier.

With the equation obtained the procedure of evaluating the temperature behaviour of the signal-to-noise ratio in an assembly of single-domain particles becomes straightforward and does not need any additional approximations (adiabaticity, high barrier). The explicit dependences of the SR curve on the essential material parameters are readily obtained. Further simplification is available with the use of an interpolation formula for the eigenvalue $\lambda_1(\sigma)$.

In a non-equilibrium situation the fluctuation–dissipation theorem fails, and the direct connection between the spectral density and susceptibility is severed. Due to this no susceptibility measurements on the system already subjected to modulation can yield the data necessary to observe SR directly.

Appendix

To prove the dominating role of the relaxation time τ_1 in the longitudinal motion of the particle magnetic moment, we shall use the effective relaxation time method. An extensive discussion of this and related problems may be found in [16].

The rotary diffusion (Fokker–Planck) equation for the distribution function $W(e, t)$ of the unit vector of the particle magnetic moment may be written [9, 10] as

$$2\sigma\tau_0\,\partial W/\partial t = \widehat{J}\,W\,\widehat{J}\,(U/k_BT + \ln W) \tag{A1}$$

where $\widehat{J}$ is the operator of infinitesimal rotations with respect to the components of e, and the energy function U is defined by equation (1). For the longitudinal component of the magnetic moment $\langle e\rangle$, equation (A1) is equivalent to the infinite set of equations for the macroscopic variables

$$\frac{2\sigma\tau_0}{l(l+1)}\frac{\mathrm{d}}{\mathrm{d}t}\langle P_l\rangle + \langle P_l\rangle - \frac{\xi}{2l+1}\left[\langle P_{l-1}\rangle - \langle P_{l+1}\rangle\right] - 2\sigma\left[\frac{l-1}{(2l-1)(2l+1)}\langle P_{l-2}\rangle + \frac{l}{(2l-1)(2l+3)}\langle P_l\rangle - \frac{l+2}{(2l+1)(2l+3)}\langle P_{l+2}\rangle\right] = 0 \tag{A2}$$

where $\xi = \mu H/k_BT$ and $\langle P_l\rangle$ is the lth Legendre polynomial of $\cos\vartheta = en$ averaged with the non-equilibrium distribution function given by equation (A1). The exact solution of the set (A2) may be presented in the form

$$\langle P_l\rangle = \sum_{k=0}^{\infty} a_{lk}\exp(-\lambda_k t/2\tau_0\sigma)$$

where the a_{lk} are the weight coefficients corresponding to the eigenvalues λ_k of equation (A1).

Now we suppose that a small steady magnetic field, formerly applied to the system, has been switched off at the moment $t = 0$. Under these circumstances the distribution function W begins to change approaching its equilibrium value at $H = 0$, i.e.,

$$W_0 = \exp(\sigma \cos^2 \vartheta)/4\pi Z(\sigma) \tag{A3}$$

where

$$Z(\sigma) = \int_0^1 \exp(\sigma y^2)\,\mathrm{d}y \tag{A4}$$

is the configurational partition function of a magnetic moment in the uniaxial anisotropy field.

The effective time of the longitudinal relaxation of the particle magnetic moment, taking into account the contributions of all the time-dependent modes, is conventionally defined as

$$\tau_{\text{eff}} = \int_0^\infty \langle P_1 \rangle \,\mathrm{d}t \Big/ \langle P_1 \rangle \Big|_{t=0} = 2\sigma\tau_0 \sum_{k=0}^{\infty} (a_k/\lambda_k) \Big/ \sum_{k=0}^{\infty} a_k\,. \tag{A5}$$

In view of definition (A5), it is convenient to rewrite the set (A2) integrating it over time. Setting there $\xi = 0$ (relaxation to the field-free state), one obtains

$$F_l - 2\sigma \left[\frac{l-1}{(2l-1)(2l+1)} F_{l-2} + \frac{l}{(2l-1)(2l+3)} F_l - \frac{l+2}{(2l+1)(2l+3)} F_{l+2} \right] = \frac{2\sigma\tau_0}{l(l+1)} \langle P_l \rangle \Big|_{t=0}. \tag{A6}$$

Here we have introduced a notation

$$F_l = \int_0^\infty \langle P_l \rangle \,\mathrm{d}t\,. \tag{A7}$$

The right-hand side (the 'initial condition') of equation (A6) for small ξ may be written as

$$\langle P_l \rangle \Big|_{t=0} = \xi \langle P_l P_1 \rangle_0 \tag{A8}$$

where the subscript 0 denotes the averaging over the equilibrium distribution (A3). With the same accuracy, equation (A7) may be presented in the form

$$F_l = \xi f_l \tag{A9}$$

where now all f_l do not depend upon ξ.

The set (A6) is easily solved numerically by the continuous fraction method [17]. The effective relaxation time (A5) is then expressed (see equations (A8) and (A9)) as

$$\tau_{\text{eff}}(\sigma) = F_1/\langle P_1 \rangle \big|_{t=0} = f_1/\langle P_1^2 \rangle_0\,. \tag{A10}$$

Note that with allowance for relation (A4) the quantity $\langle P_1^2 \rangle_0$ coincides with the parameter B entering equation (4).

The numerical results of evaluation of $\tau_{\text{eff}}(\sigma)$ by the described procedure are shown in table A1 in comparison with $\tau_1(\sigma)$. The high accuracy of their closeness is apparent, and this justifies the use of τ_1 in equation (4).

Table A1. Comparison of the effective and single-mode values of the longitudinal relaxation time.

σ	$\tau_{eff}/\sigma\tau_0$	$2/\lambda_1$
10^{-3}	1.000 40	1.000 40
10^{-2}	1.004 01	1.004 01
10^{-1}	1.041 05	1.041 07
1	1.527 98	1.531 07
5	14.588 8	14.770 4
10	691.016	693.922
25	$5.322\,69\times10^8$	$5.325\,23\times10^8$

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