Stochastic resonance and phase shifts in superparamagnetic particles

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In single-domain particles magnetic moments behave as overdamped rotary oscillators with a potential $\propto \cos^2 \theta$. Thus they make a well-understandable object for studies of stochastic resonance (SR). With a numerically exact approach, we analyze magnetic SR in superparamagnetic assemblies including a situation when an external field creates a controllable inequality of the potential-well depths.

INTRODUCTION

In recent papers¹⁻⁷ on stochastic resonance (SR), the main model under investigation is an overdamped anharmonic oscillator with the potential $U(q) = -aq^2 + bq^4$, both a and b being positive. Though possible to realize experimentally,² this model does not at all exhaust the variety of systems displaying the SR behavior. For example, the importance of studies of magnetic SR had been clearly outlined in Ref. 8. Hereby we present a consideration of SR in ^a well-known material object—an assembly of single-domain particles embedded in a solid nonmagnetic matrix. With some minor changes it is valid also for magnetic fluids-suspensions of anisotropic ferroparticles.

Single-domain particles of a ferromagnet or ferrite with a uniaxial anisotropy provide a perfectly understandable and simple in realization example of a bistable system sensitive to the thermal noise. As it had been shown experimentally yet in the middle of the 1950s (Ref. 9) and since then many times verified (see Ref. 10, for example), with the particles of a size $\sim 100 \text{ Å}$ in the temperature range about 100K, one may easily proceed from the conventional ferromagnetism (particles being tiny permanent magnets) to *superparamagnetism*, i.e., intense fluctuational motion of a magnetic moment inside the particle. The parameter responsible for the change of regimes is $\sigma = KV/k_BT$ —the ratio of the magnetic anisotropy barrier $(K$ is the first anisotropy constant and V the particle volume) to the thermal energy k_BT .

Since its foundation by $N\acute{e}el^{11}$ in 1949, the theory of relaxation processes in single-domain particles had undergone a considerable development.¹²⁻¹⁸ Here we employ it to analyze SR.

The below presented results give an account and a set of conventional characteristics (signal-to-noise ratio, phase shift) of SR in a rotationally bistable system with the potentials $U(\vartheta) = -a \cos^2 \vartheta$ or $-a \cos^2 \vartheta - b \cos \vartheta$ which correspond to the cases of zero and finite bias magnetic field, respectively. We deal in the framework of the linear-response theory specified for SR in Refs. 1 and 3, i.e., express the results in terms of the dynamic susceptibility χ . However, unlike former attempts, ^{19,20} while evaluating χ , we retain a big enough part of the relaxation spectrum, thus making our calculations numerically exact. In other words, all the significant contributions of intrawell modes are always taken into account. In this aspect our paper is closely related to Ref. 21, where the same potential has been analyzed numerically in the case of color-noise driven stochastic relaxation.

I. GENERAL RELATIONS

Consider a uniformly magnetized (single-domain) particle with a magnetic moment $\mu = \mu e$, where e is a unit vector. The particle has a uniaxial magnetic anisotropy (crystalline or other) with the energy density K ; the direction of the anisotropy axis is denoted by a unit vector n. let the particle (or an assembly of identical particles) be fixed inside some nonmagnetic solid matrix. In the case of an assembly, we assume that the particle concentration is small enough as to be able to neglect their magnetic dipole-dipole interaction.

The orientation-dependent part of the particle energy in the absence of external magnetic fields is

$$
U = -KV(\mathbf{en})^2 \,, \tag{1}
$$

where the variable $(en) = \cos \vartheta$ is in fact the normalized projection of the magnetic moment on the direction of the anisotropy axis. In the presence of a thermal bath, the orientational distribution function $W(e, t)$ of vector e obeys the Fokker-Planck-like equation^{12,1'}

$$
\frac{\partial}{\partial t}W + \widehat{\Lambda}W = 0, \quad \widehat{\Lambda} = -\frac{1}{2\sigma\tau_0} \widehat{J} \left(\frac{1}{k_B T} W \widehat{J} U + \widehat{J}W \right), \tag{2}
$$

where τ_0 is the decay time of the Larmor precession in a bulk material (a dimensionless rate of spin-lattice relaxation), and $\hat{\mathbf{J}} = (\mathbf{e} \times \partial/\partial \mathbf{e})$ is the operator of an infinitesimal rotation. The stationary solution of Eq. (2) reads

$$
W_0 = Z_0^{-1} \exp(\sigma \cos^2 \vartheta), \quad Z_0 = 4\pi G(\sigma),
$$

$$
G(\sigma) = \int_0^1 \exp(\sigma x^2) dx.
$$
 (3)

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The stochastic resonance is determined by the longitudinal (with respect to $n)$ modes of the relaxational problem (2) . Since Λ is not a self-adjoint operator, it produces, together with the spectrum of eigenvalues $\{\lambda_i\},\$ two sets of eigenfunctions defined as

$$
\widehat{\Lambda}\varphi_i=\lambda_i\varphi_i\,,\qquad \widehat{\Lambda}^+\psi_j=\lambda_j\psi_j\,,
$$

where + denotes the Hermitian conjugation. These eigenfunctions are connected by relation

$$
\varphi_i = W_0 \psi_i , \qquad (4)
$$

and orthonormalized. Their expansion in the Legendre polynomial series

$$
\varphi_i = \frac{1}{2} \sum_{l=0}^{\infty} (2l+1) a_l^{(i)} P_l(\cos \vartheta), \quad \psi_j = \sum_{l=0}^{\infty} b_l^{(j)} P_l(\cos \vartheta), \tag{5}
$$

produces two sets of eigenvectors, ${a_l}^{(i)}$ and ${b_l}^{(j)}$, the components of which, as follows from Eqs. (4) and (5), are related to each other by

$$
a_l^{(k)} = [\langle P_l P_{l'} \rangle_0 - \langle P_l \rangle_0 \langle P_{l'} \rangle_0] b_{l'}^{(k)},
$$

where the subscript 0 denotes the averaging over the equilibrium distribution W_0 from Eq. (3).

The Green function of Eq. (2), i.e., the probability density of a state (x, t) , providing the initial state is $(x_0, 0)$, reads

$$
W(x,t|x_0) = \sum_{k=0}^{\infty} \varphi_k(x) \psi_k(x_0) e^{-\lambda_k t}; \qquad (6)
$$

hereafter we use the notation $x = \cos \vartheta$. The subject of our interest is the correlator $\mu^2 \langle \langle x(t)x(0) \rangle \rangle_0$, where the averaging over $x(t)$ is performed with the function W from Eq. (6) , and that over the initial conditions—with the equilibrium function W_0 . Using formulas (4)–(6), one gets

$$
\mu^{2} \langle \langle x(t)x(0) \rangle \rangle_{0} = \mu^{2} \int \int dx dx_{0} x x_{0} W(x, t|x_{0}) W_{0}
$$

$$
= \mu^{2} \sum_{k=1}^{\infty} \left[a_{1}^{(k)} \right]^{2} e^{-\lambda_{k} t}; \tag{7}
$$

the summation here is taken only over the odd values of k.

Transforming the correlator (7) by the Kubo formula, one arrives at the longitudinal dynamic susceptibility

$$
\chi(\Omega) = \chi' - i\chi'' = (\mu^2 B/k_B T) \sum_{k=1} w_k \lambda_k / (\lambda_k + i\Omega),
$$

$$
B = [(\cos^2 \vartheta)_0 - (\cos \vartheta)_0^2], \qquad (8)
$$

of a single-domain particle with respect to the external field $H(t) = H \exp(i\Omega t)$; here we have introduced a notation $w_k = B^{-1} \left[a_1^{(k)} \right]^2$ so as $\sum w_k = 1$.

According to the fluctuational-dissipation theorem, the spectral density function in terms of χ reads¹

$$
Q(\omega) = \pi H^2 |\chi|^2 \delta(\omega - \Omega) + 4 \frac{k_B T}{\omega} \chi''(\omega). \tag{9}
$$

Setting $\omega = \Omega$ (the necessary condition of the onset of SR) and comparing the signal-induced $(\delta$ -function) and noise (proportional to T) contributions in expression (9) , one gets with the aid of Eq. (8) the signal-to-noise ratio **as**

$$
S = \frac{\pi}{4\tau_0} \left(\frac{IH}{K}\right)^2 R(\sigma, \Omega),
$$

\n
$$
R(\sigma, \Omega) = \sigma^2 B \left\{ \left[\sum \frac{w_k}{1 + (\Omega \tau_k)^2} \right]^2 + \left[\sum \frac{\Omega w_k \tau_k}{1 + (\Omega \tau_k)^2} \right]^2 \right\} / \sum \frac{w_k \tau_k / \tau_0}{1 + (\Omega \tau_k)^2},
$$
\n
$$
(10)
$$

and the phase shift

$$
\phi(\sigma,\,\Omega) = -\arctan\left(\chi''/\chi'\right) = -\arctan\left[\sum \frac{\Omega w_k \tau_k}{1 + (\Omega \tau_k)^2} / \sum \frac{w_k}{1 + (\Omega \tau_k)^2}\right],\tag{11}
$$

where we have introduced the spectrum of relaxation times $\tau_k = 1/\lambda_k$, and made use of the relation $\mu = IV$, where I is the magnetization of a ferromagnet, valid for single-domain particles. As long as summations in Eqs. (10) and (11) are infinite, the corresponding expressions represent exact results for SR in a single-domain particle assembly.

II. MAGNETIC STOCHASTIC RESONANCE

The sets of relaxation times τ_k and weight coefficients w_k were evaluated numerically. Substitution of expansions (5) into the Fokker-Planck equation (2) yields a homogeneous tridiagonal recurrence relation

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$$
\left[1 - \frac{\lambda_i}{l(l+1)}\right] a_l^{(i)} - 2\sigma \left[\frac{l-1}{(2l-1)(2l+1)} a_{l-2}^{(i)} + \frac{1}{(2l-1)(2l+3)} a_l^{(i)} - \frac{l+2}{(2l+1)(2l+3)} a_{l+2}^{(i)}\right] = 0,
$$
\n(12)

and the one corresponding to the conjugate equation reads

$$
[l(l+1)-\lambda_i] b_l^{(i)} + 2\sigma \left[\frac{(l-2)(l-1)l}{(2l-3)(2l-1)} b_{l-2}^{(i)} - \frac{l(l+1)}{(2l-1)(2l+3)} b_l^{(i)} - \frac{(l+1)(l+2)(l+3)}{(2l+3)(2l+5)} b_{l+2}^{(i)} \right] = 0. \tag{13}
$$

For each *i* the eigenvalue λ_i and the sets of corresponding eigenvectors $\{a_l\}^{(i)}$ and $\{b_l\}^{(j)}$ have been found with the aid of the continued fraction algorithm.²²

Our calculations show that to achieve good accuracy with Eqs. (10) and (11) in a wide range of temperature and frequency variations, it is necessary to retain at least five (odd $k = 1, 3, ..., 9$) lower modes of the spectrum. We remark that the first three relaxational modes have been once evaluated both numerically²³ and analytically²⁴ in studies of dielectric relaxation in nematic liquid crystals, where the forms of the basic equation and potential coincide with those given by our Eqs. (2) and (1) , respectively.

The obtained dependences of S on the temperature parameter $\sigma^{-1} = k_B T/KV$ are shown in Figs. 1 and 2. Note that within the framework of the linear response theory, our approach removes all the restrictions on the frequency range, i.e., the SR characteristics are evaluated correctly for any Ω .

As it must have been expected, the SR, that is the main maximum of $R(\sigma)$ at $\sigma \sim 1$, is most pronounced at $\Omega \rightarrow 0$ and zero bias field—see curve 1 of Fig. 1. This case is relatively simple, since in the $\Omega = 0$ limit Eq. (10) reduces to

$$
R(\sigma) = \sigma^2 B \tau_0 / \tau_{\text{eff}} , \qquad \tau_{\text{eff}} = \sum_{1}^{\infty} w_k \tau_k . \qquad (14)
$$

FIG. 1. Signal-to-noise ratio as a function of a dimensionless temperature. For $\Omega \tau_0 = 0$ see dotted curve at the bottom of the minimum of curve 1. Solid curves: $\Omega \tau_0 = 0.01$ (1), 0.1 $(2), 0.5 (3), 1 (4), 2 (5), 10 (6).$

The last expression justifies the treatment of magnetic SR in the $\Omega = 0$ limit as a process characterized by just one effective relaxation time τ_{eff} . As it has been shown in Refs. 18 and 20, the time τ_{eff} is very close to τ_1 in a wide range of σ .

However, the case of Ω being exact zero is to some extent an exception. At any finite Ω one has to remember that the time τ_1 is exponential in σ , i.e., grows infinitely with cooling the system. Due to that at low temperatures the interwell transition is completely "frozen," and the situation is governed by intrawell relaxation. The latter is sensitive to the details of the potential near the bottom of the well, and for the system in question is determined by the infinite eigenvalue spectrum λ_k of the kinetic equation (2) for $k > 3$.

The low-temperature limit of the function $R(\sigma)$ may be derived with the aid of asymptotic expressions for χ given in Ref. 24,

$$
\chi = \frac{\mu^2}{k_B T} \left[\left(1 - \frac{1}{\sigma} - \frac{3}{4\sigma^2} \right) \frac{1}{1 + i\Omega \tau_1} + \frac{1}{8\sigma^2} \left(\frac{1}{1 + i\Omega \tau_3} + \frac{1}{1 + i\Omega \tau_5} \right) + O(\sigma^{-3}) \right], \qquad (15)
$$

where $\tau_1 = \tau_0 \sqrt{\pi/4 \sigma} e^{\sigma}$ and $\tau_3 = \tau_5 = \frac{1}{2} \tau_0 (1 + 5/2 \sigma + ...)$.

FIG. 2. Signal-to-noise ratio at nonzero values of the applied constant field $\xi = \varepsilon \sigma$ for $\varepsilon = 0.1$ (1), 0.2 (2), 0.5 (3), $1 \; (4)$, $2 \; (5)$, $5 \; (6)$. All the results correspond to the limit $\Omega\tau_0=0.$

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The first term of this expansion describes the interwell relaxation process responsible for SR proper-the right maximum in Fig. 1. But at low temperatures $\sigma \gg 1$ and due to that τ_1 is exponentially large. Because of that one has to set $\Omega \tau_1 \gg 1$ for any finite frequency, and the first term drops out. The second and following terms in Eq. (15) render the contribution of intrawell relaxation whose rate remains finite at $T \to 0$. Substituting the truncated Eq. (15) into Eqs. (9) and (10) , instead of zero predicted by formula (14) one obtains $R(\sigma \to \infty) = \frac{1}{2}$. Yielding the correct limiting value, Eq. (15) however, is not accurate enough for a correct description of $R(\sigma)$ at large σ , i.e., low temperatures. Indeed, it is easy to show that it gives $(dR/d\sigma^{-1})_0$ negative whereas the exact numerical treatment proves that the initial temperature
slope of the $R(1/\sigma)$ curve is positive—see Fig. 1. This slope of the $R(1/\sigma)$ curve is positive—see Fig. 1. This low-temperature increase of the function $R(1/\sigma)$ causes the additional maximum at the signal-to-noise (SR) ratio in superparamagnetic systems.

With minor complications our method provides a di-

rect way to study the case of nonequal potential wells. Physically it means merely that some bias constant field H_0 is applied to the particle parallel to its anisotropy axis n . Then the energy function (1) is replaced by

$$
U=-\mu H_0(\boldsymbol{e}\boldsymbol{n})-KV(\boldsymbol{e}\boldsymbol{n})^2\,,\qquad\qquad(16)
$$

where now the first term \propto cos ϑ breaks the bidirectional symmetry of the potential. However, the two-minima pattern for the potential $U(\vartheta)$ exist as long as the bias field is smaller than $H_c = 2K/I$ —the maximum coercive force of a single-domain particle. In the range $H_0 < H_c$, though the wells' equilibrium populations and transition rates are different, the magnetic moment still undergoes interwell motions.

For this case the Fokker-Planck equation (2) redefined with regard to Eq. (16) after the procedure similar to that yielding Eqs. (12) and (13) turns into a pentadiagonal recurrence relationship

$$
\left[1 - \frac{\lambda_i}{l(l+1)}\right] a_l^{(i)} - \frac{\xi}{2l+1} \left[a_{l-1}^{(i)} - a_{l+1}^{(i)}\right]
$$
\n
$$
-2\sigma \left[\frac{l-1}{(2l-1)(2l+1)} a_{l-2}^{(i)} + \frac{1}{(2l-1)(2l+3)} a_l^{(i)} - \frac{l+2}{(2l+1)(2l+3)} a_{l+2}^{(i)}\right] = 0.
$$
\n(17)

The conjugated one reads

$$
[l(l+1) - \lambda_i] b_l^{(i)} + \xi \left[\frac{(l-1)l}{2l-1} b_{l-1}^{(i)} - \frac{(l+1)(l+2)}{2l+3} b_{l+1}^{(i)} \right]
$$

+2\sigma \left[\frac{(l-2)(l-1)l}{(2l-3)(2l-1)} b_{l-2}^{(i)} - \frac{l(l+1)}{(2l-1)(2l+3)} b_l^{(i)} - \frac{(l+1)(l+2)(l+3)}{(2l+3)(2l+5)} b_{l+2}^{(i)} \right] = 0. (18)

These recurrence relations are solved using matrix continued fraction method.^{21,22} Figure 2 shows how the decay of SR takes place due to gradual shoaling of one of the minima under growth of the bias field H_0 . To characterize the latter, in Eqs. (17) and (18) we have introduced a dimensionless parameter $\xi = \mu H_0/k_BT$. With such a choice, the ratio $\varepsilon = \xi/\sigma$ does not depend upon temperature and gives the strength of the external field in the units of the internal (i.e., anisotropy) one.

As the field strength grows, the position of the SR maximum moves rightward, to higher temperatures. Normally, one would have expected a shift to the opposite side because of the increase of the net relaxation rate. To explain the "reversed" shift one has to note that the presence of the field cardinally changes the temperature behavior of the coefficient B —the static susceptibility of the system. At $H_0 = 0$, when $\langle \cos \vartheta \rangle_0 = 0$, with the $\text{temperature growth it } \textit{diminishes} \text{ from 1 to } \frac{1}{3} \text{ rendering}$ the initial susceptibility. However, under nonzero field the effect of saturation of the longitudinal magnetization, yielding $B \to 0$ at $T \to 0$ becomes essential, and B

grows from zero at $T \to 0$ to $\frac{1}{3}$ at $T \to \infty$. Due to that at $H_0 \neq 0$ the combination $\sigma^2 B$ [see Eq. (10)] acquires a temperature maximum of its own. It is this specific (static) temperature-dependent factor that reverses the direction of the SR maxima shift.

III. PHASE BEHAVIOR,

The developed approach is very convenient to obtain a comprehensive account of the frequency and temperature behavior of the phase shift in the system in question. The problem has special interest since those dependences for bistable systems have been put under discussion in Refs. 5 and 6 with contradicting conclusions. Some particular numerical simulations on the same subject reported in Ref. 7, though interesting, are insufficient to draw out a final clarification.

Before proceeding to discussion of the details of the phase behavior, we would like to emphasize after the authors of Ref. 25 that neither lack nor presence of the

phase maximum may be taken as a "signature" of the SR proper. Actually, the maxima of R and $|\phi|$ have essentially different origin. Whereas the first one occurs only under certain match between the interwell hopping rate and the external frequency, the second depends just on the very existence of intrawell transitions. A large difference in the positions of these maxima (cf. Figs. 1 and 3) and the existence of the $|\phi|$ peak in the range $\Omega \tau_0 \gg 1$ where SR is clearly absent (cf. curves $5, 6$ in Fig. 1 and curves 2,8 in Fig. 4), are the particular manifestations of this fact. However, despite that there is no such a thing as a stochastic resonance of the phase shift, the behavior of $|\phi|$ under SR conditions is definitely worth consideration.

The main point of the argument set out in Refs. 5 and 6 is whether $|\phi|$ increases or decreases in the lowtemperature limit at high frequencies, i.e., $\Omega \tau_0 \geq 1$. Note that the reversed characteristic intrawell time τ_0^{-1} which remains finite at $T \rightarrow 0$ is the only natural frequency scaling parameter here, since the interwell hopping rate τ_1^{-1} , being exponential in T, may not be used for this purpose.

The temperature-frequency behavior of the phase shift in magnetic SR evaluated by rigorous numerical procedure is presented in Fig. 3 and with special emphasis to high frequencies in Fig. 4. The asymptotic representation (15) helps in understanding the origin of the limit $\phi(0) = \phi(T \to 0)$ and the form of the curves at low temperatures, i.e., at $\Omega \tau_1 \rightarrow \infty$ and $\sigma \gg 1$. Under these conditions Eq. (15) yields for the phase shift

$$
-\phi\bigg|_{1/\sigma=0} = \arctan\left[\frac{1}{2}\Omega\tau_0\left(1+\frac{5}{2\sigma}\right)\right],\qquad(19)
$$

which is in a full agreement with the numerical data of Figs. 3 and 4.

Differentiating Eq. (19) with respect to $1/\sigma$, that is

FIG. 3. Phase shift as a function of a dimensionless temperature for $\Omega \tau_0 = 0.01$ (1), 0.1 (2), 0.5 (3).

FIG. 4. Phase shift as a function of dimensionless temperature at high frequencies; $\Omega \tau_0 = 1$ (1), 2 (2), 10 (3), 50 (4).

dimensionless temperature, one gets for the initial slope $\left| \phi(T) \right| \text{ curve}$

$$
\left. \frac{d|\phi|}{d\sigma^{-1}} \right|_{1/\sigma=0} = \frac{5\Omega\tau_0}{4\left(1 + \frac{1}{4}\Omega^2\tau_0^2\right)}.
$$
 (20)

That means that at any finite frequency the absolute value of the phase always increases with temperature. This result is consistent with the simulations of Ref. 6. Apparently, the effect is entirely due to intrawell motions and plays the main role in the temperature range where the interwell process may be neglected. But as soon as

FIG. 5. Position of the maximum of the function $|\phi(T)|$ against dimensionless frequency.

the latter is activated, it immediately becomes the dominating one. Together with creating conditions for SR, the interwell process tends function $|\phi(T)|$ down to zero. Given that, Eq. (20) is a direct proof of existence of a maximum at the temperature dependence whatever large Ω may be.

An overview of the curves in Figs. 3 and 4 suggests that at $\Omega \tau_0 \gg 1$ the position $(1/\sigma)_m$ of this maximum rather weakly if ever depends upon Ω . Numerical investigation confirms this conclusion. As it is shown in Fig. 5, the value of $(1/\sigma)_{\rm m}$ first grows rapidly, then passes through a maximum, and finally descends very slowly to the limit $(1/\sigma)_m = 0$ at $\Omega \to \infty$.

Dealing in terms of intra- and interwell transitions, it is easy to understand also the conclusion of Ref. 5 prescribing a monotonous decrease and no maximum of $|\phi(T)|$. Though wrong for our case, it is valid for a system of a special type—the one which completely lacks any intrawell degrees of freedom. For such a model, instead of formulas (11) or (15), the phase is exhaustively described by a relationship $\phi = -\arctan \Omega \tau_1$. Then for any Ω , however small, at $T \rightarrow 0$ it tends to $-\pi/2$ because of the fast growth of the response time. Any heating causes reduction of τ_1 and hence diminution of $|\phi|$. Comparison of the two cases proves that with respect to the phase behavior, the systems with or without intrawell processes, though both capable of SR, are qualitatively different.

CONCLUSIONS

Let us sum up the presented theoretical evidence.

Single-domain particle assemblies offer a wellrealizable possibility to study all the scope of efFects pertinent to SR.

The kinetic (rotary diffusion) equation for the particle magnetic moment may be easily solved with high precision thus taking into account contributions from the intrawell motions which are essential for a correct description of SR, especially at low temperatures.

This kinetic model does not use any kind of adiabatic assumption, and due to that is capable to render frequency dependences of the SR characteristics.

By application of the external constant field of arbitrary strength, the symmetrical double-well potential may be gradually transformed into a single-well (dipolar) one; upon growth of the bias field, SR maximum noticeably shifts to higher temperatures.

In the low-temperature —limit the phase of magnetic oscillations $|\phi|$ increases with temperature due to intrawell relaxation processes.

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