Precession-aided magnetic stochastic resonance in ferromagnetic nanoparticles with cubic anisotropy

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It is shown that the signal-to-noise ratio in the magnetic stochastic resonance of single-domain ferromagnetic nanoparticles having cubic anisotropy exhibits a strong intrinsic dependence on the decay rate α of the Larmor precession. This dependence (precession aided relaxation) is due to coupling between longitudinal relaxation and transverse (precessional) modes arising from the lack of axial symmetry. It is most pronounced in the intermediate-to-low damping (Kramers turnover) region $0.1 < \alpha < 1$. The effect which does not exist for axially symmetric potentials may be used to determine α.

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Stochastic resonance (SR) is nowadays a well known but still remarkable effect which allows one to control the behavior of periodic signals passing through noisy systems. As a manifestation of cross-coupling between stochastic and regular motions, the SR effect is universal in physics (e.g., optics, mechanics of solids, superconductivity, surface science), communications engineering (optimal detection and tracing of signals) as well as in various branches of chemistry and biology. Comprehensive reviews of diverse aspects of SR are available in Refs. 1–3.

The archetypal theoretical model of SR[1] is a heavily damped (so that inertial effects can be ignored) Brownian particle in a bistable potential subjected to noise arising from a thermal bath. The particle is excited by an ac driving force of frequency Ω close to the rate of Kramers transitions (escape rates) between the wells nevertheless with amplitude insufficient to induce the transitions. Consequently, switching may occur only by the combined effect of the regular ac force and the noise. The spectral density $\Phi(\omega)$ of the motion at the frequency $\omega = \Omega$ is then evaluated, and the resulting signal-to-niose ratio (SNR) (or the spectral power amplification coefficient) is analyzed as a function of the noise intensity D. The curve SNR(D) has a bell-like shape, i.e., it passes through a maximum thus exhibiting *stochastic resonance*. The maximum in the SNR is interpreted as due to the remarkable ability of noise to enhance the intensity of the interwell hoppings in the system.

Superparamagnetism of fine particles subject to a weak ac field is yet another important manifestation of SR. Here the magnetic anisotropy provides the multistable states for the magnetization **M** of the particle while the thermal fluctuations due to the bath which is in perpetual thermal equilibrium at temperature T are the source of the noise. These conditions give rise to *magnetic stochastic resonance* which may be defined as the enhancement of the SNR due to noise.[4] The superparamagnetism of single-domain ferromagnetic nanoparticles until recently entirely of academic interest, is now important in technology because of the ever decreasing size of the particles used in magnetic recording. Once the size of the particle is reduced to nanoscale dimensions superparamagnetism imposes a physical limit on the miniaturization of the bit-recording elements. Hitherto in the context of micromagnetism the size of the particles used ensured that thermal effects occurring at room temperature could safely be ignored. However, due to ongoing miniaturization, a superparamagnetic nanoparticle is now an acceptable model for both writable elements and read/write heads.[5]

The magnetic SR phenomenon is clearly manifested by *uniaxial* single-domain particles.[6–9] Here the magnetic free energy density V is governed by a single coordinate ϑ (the colatitude) and is given by

$$\beta V(\vartheta) = \Xi \sin^2 \vartheta, \tag{1}$$

where $\beta = v/kT$, v is the volume of the particle, k is Boltzmann's constant, $\Xi = \beta K_u$ is the dimensionless barrier height parameter, and K_u is the anisotropy constant. In the absence of external magnetic fields, the magnetization of the uniaxial particle has two equivalent stable orientations at $\vartheta = 0$ and $\vartheta = \pi$, so that it is an ideal example of a bistable system subjected to noise. Magnetic SR in grains possessing uniaxial anisotropy (characterized by the single coordinate ϑ) has been treated in Refs. 6–11 and is completely understood. In contrast, we have little knowledge about magnetic SR in nonaxially symmetric potentials governed by *two* coordinates ϑ and φ (φ is the azimuthal angle) which in general give rise to *multistable* states. For illustration, consider a *cubic* crystal, where the particle energy is[12]

$$\beta V(\vartheta, \varphi) = \sigma(\sin^4 \vartheta \sin^2 2\varphi + \sin^2 2\vartheta), \tag{2}$$

$\sigma = \beta K_c / 4$ and K_c is the anisotropy constant, which may be either positive or negative. For $K_c > 0$ (Fe-type crystals) the potential (2) has six minima (wells), eight maxima, and 12 saddle points [see Fig. 1(a)]. For $K_c < 0$ (Ni-type crystals) the maxima and minima interchange their roles while the saddle points do not alter [Fig. 1(b)].

Here we demonstrate that magnetic SR in particles with cubic anisotropy has behavior which has little in common

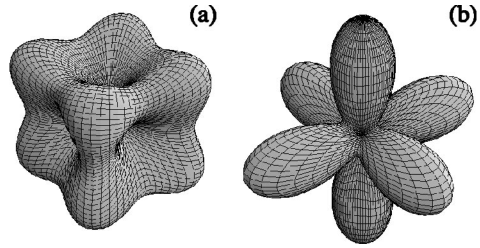

FIG. 1. Cubic anisotropy potential for $K_c > 0$ (a) and $K_c < 0$ (b).

with that for uniaxial anisotropy due to the lowered local symmetry (both ϑ and φ are involved) of the energyscape in the vicinity of the easy axis direction in the space of orientations. In uniaxial anisotropy, the energyscape is a uniform equatorial ridge (zone) separating two polar minima and has no saddle points, on the other hand, cubic anisotropy generates azimuthally nonuniform energy distributions with numerous saddle points. Such a nonaxially symmetric energyscape leads to a new effect, viz., strong intrinsic dependence of SR in superparamagnetic particles with cubic anisotropy on the rate of spin-lattice relaxation which is determined by the damping coefficient α. To understand qualitatively this effect, we consider the rotational motion of the magnetization of the nanoparticle at zero temperature (no fluctuations) which is described by the Landau–Lifshitz–Gilbert equation

$$\dot{\mathbf{e}} = [\mathbf{e} \times (\gamma \mathbf{H}_{\text{eff}} - \alpha \dot{\mathbf{e}})], \tag{3}$$

where $\mathbf{e} = \mathbf{M}/M_S$ (M_S is the saturation magnetization), $\mathbf{H}_{\text{eff}} = -\partial V/\partial \mathbf{M}$ and γ is the gyromagnetic ratio. To induce SR in such a particle, a weak spatially uniform ac magnetic field $\mathbf{H}$ of frequency Ω favoring the Kramers hopping of the magnetization between its equilibrium positions is imposed along its easy axis $\mathbf{n}$. For uniaxial anisotropy, the magnetic SNR does not intrinsically depend on α.[8] Conversely, in nonaxially symmetric potentials, e.g., cubic anisotropy, the situation is entirely different. First, the presence of several easy axes orthogonal to each other means that the notion of longitudinal and transverse orientations (with respect to $\mathbf{n}$) has no meaning. Second, as one may deduce from Eq. (3), in the "folded" energyscape, the friction torque acting on $\mathbf{M}$ is no longer directed at a constant angle to the direction of precession thus strongly modulating the angular velocity of precession. Hence, reorientation of $\mathbf{M}$ has a strong intrinsic dependence on α, i.e., we have *precession aided relaxation*. Apparently, precession aided relaxation is a universal feature of particles with nonaxially symmetric potentials.[13]

Magnetic SR may be generally described using linear response theory as follows.[14–16] The Fourier components of the magnetization of the particle M_ω and of the applied ac field H_ω are related via $M_\omega = \chi(\omega) H_\omega$, where $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ is the complex magnetic susceptibility of the particle. The noise-induced part $\Phi_M^{(n)}(\Omega)$ is obtained using the fluctuation-dissipation theorem as $\Phi_M^{(n)}(\Omega) = kT\chi''(\Omega)/(\pi v \Omega)$. The spectral density $\Phi_M^{(s)}(\Omega)$ of the forced magnetic oscillations in a field $H(t) = H\cos\Omega t$ at the excitation frequency Ω is

$$\Phi_M^{(s)}(\Omega) = \frac{1}{2} \lim_{\Delta\Omega \to 0} \int_{\Omega-\Delta\Omega}^{\Omega+\Delta\Omega} (H|\chi(\omega)|)^2 \times [\delta(\omega+\Omega) + \delta(\omega-\Omega)] d\omega,$$

where the parity condition $\chi^*(\omega) = \chi(-\omega)$ has been used. Combining the above equations, one obtains[16]

$$\text{SNR} = \frac{\Phi_M^{(s)}}{\Phi_M^{(n)}} = \frac{\pi \Omega H^2 v |\chi(\Omega)|^2}{2kT\chi''(\Omega)}. \tag{4}$$

The susceptibility is given in the adiabatic limit $\Omega \to 0$,[16]

$$\chi(\Omega) \approx \chi_0 (1 - i\Omega \tau_c), \tag{5}$$

where $\chi_0 = \beta M_S^2/3$ is the static susceptibility and τ_c is the correlation time defined as the area under the curve of the equilibrium correlation function of the projection of $\mathbf{M}$ in the direction of the exciting field, viz.,

$$\tau_c = \int_0^\infty \frac{\langle \cos\vartheta(t) \cos\vartheta(0) \rangle_0}{\langle \cos^2 \vartheta(0) \rangle_0} dt, \tag{6}$$

and the subscript zero denotes the equilibrium statistical average over the realizations of $\cos\vartheta$. Here we have used the relation $\langle \cos^2\vartheta \rangle_0 = 1/3$ valid for cubic crystals. Substituting Eq. (5) into Eq. (4) and taking the limit $\Omega \to 0$, we have

$$\text{SNR} = 2\pi\omega_K (M_S H/K_c)^2 R(\sigma,\alpha)/3,$$

where $\omega_K = 2\gamma K_c/M_S$ is a reference frequency of order of magnitude the Larmor precession frequency in the internal anisotropy field and the dimensionless factor R is given by

$$R(\sigma,\alpha) = |\sigma| \tau_0/\tau_c(\sigma,\alpha), \tag{7}$$

and $\tau_0 = \beta M_S/(2\gamma)$ is a reference time. The rate of noise induced transitions between the potential wells is controlled by the anisotropy parameter $\sigma = vK_c/(4kT)$ so that $|\sigma|^{-1}$ is the dimensionless temperature, i.e., the noise intensity.

The evaluation of the SNR for cubic crystals can be accomplished as follows. To describe the magnetization relaxation of a *superparamagnetic* particle (i.e., a particle with *finite* barrier height parameter), the magnetodynamic Eq. (3) must be augmented by a random field with white noise properties so becoming a Langevin equation.[17] The corresponding Fokker–Planck equation for the distribution function $W(\mathbf{e},t)$ of orientations of $\mathbf{e}$ is[17]

$$\frac{\partial}{\partial t} W = \frac{1}{2\tau_N} \{ \beta[\alpha^{-1} \mathbf{e} \cdot (\nabla V \times \nabla W) + \nabla \cdot (W \nabla V)] + \Delta W \}, \tag{8}$$

where ∇ and Δ are the gradient and Laplacian on the surface of the unit sphere, $\tau_N = \tau_0(\alpha + \alpha^{-1})$ is the characteristic free diffusion time of the magnetization. The solution of the Fokker–Planck Eq. (8) is reduced by Fourier expansion in the spherical harmonics $Y_{l,m}(\vartheta,\varphi)$ to the solution of an infinite hierarchy of differential-recurrence equations for the correlation functions $c_{l,m}(t) = \langle \cos\vartheta(0) Y_{l,m}[\vartheta(t),\varphi(t)] \rangle_0$, so that $C(t) = c_{10}(t)/c_{10}(0)$,[16]

$$\frac{d}{dt}c_{lm}(t) = \sum_{l'm'} d_{lml'm'}c_{l'm'}(t). \tag{9}$$

Here $d_{lml'm'}$ are the matrix elements of the Fokker–Planck operator in Eq. (8); these elements are listed explicitly in Refs. 16, 18, and 19. [The derivation of Eq. (9) for an arbitrary free energy function $V(\vartheta,\varphi)$ is given in Ref. 20.] The *exact solution* of Eq. (9) for the Laplace transform $\tilde{c}_{1,0}(s)$ can be obtained by matrix continued fractions.[16,18,19] Having determined $\tilde{c}_{1,0}(s)$, one may calculate $\tau_c = \tilde{c}_{10}(0)/c_{10}(0)$ (see Refs. 18 and 19 for details) and the SNR from Eq. (7).

Although the matrix continued fraction method yields the exact values of the SNR, its application is rather limited since the qualitative behavior of the SNR is not obvious by this method. However, the behavior of the SNR can readily be understood in qualitative fashion by noting that in the low temperature (high barrier) limit, $\sigma \gg 1$, the correlation times τ_c for cubic crystals may be given by simple asymptotic formulas. These asymptotic equations for τ_c were obtained in the context of the Kramers theory[21] for the thermally activated escape rate of a mechanical particle from a potential well as extended to the magnetic problem by Brown,[17] Smith and de Rozario,[22] Klik and Gunther,[23] and Coffey *et al.*[24]) The appropriate formula for τ_c valid in the very low damping (VLD, $\alpha \to 0$) and intermediate-to-high damping (IHD, ($\alpha \geqslant 1$) limits have been given in Ref. 23 and Refs. 19 and 22, respectively. Furthermore, universal formulas which apply like the exact matrix continued fraction solution to all damping regimes have been obtained in Ref. 25. Thus, we can readily estimate the SNR in the low-temperature limit, viz.,

$$R \sim R_{\mathrm{IHD}} e^{(1/\pi)\int_0^\infty \ln(1-e^{-8\sqrt{2}\alpha|\sigma|(\lambda^2+1/4)/9})[d\lambda/(\lambda^2+1/4)]}, \tag{10}$$

where R_{IHD} is the SNR for IHD values of damping ($\alpha \geqslant 1$) given by

$$R_{\mathrm{IHD}} \sim \begin{cases} \dfrac{2\sqrt{2}\sigma^2 e^{-\sigma}}{\pi(1+\alpha^2)}(\sqrt{9\alpha^2+8}+\alpha), & K_c > 0, \\ \dfrac{2\sqrt{2}\sigma^2 e^{-|\sigma|/3}}{3\pi(1+\alpha^2)}(\sqrt{9\alpha^2+8}-\alpha), & K_c < 0, \end{cases} \tag{11}$$

and the exponential factor bridges the VLD and IHD values of R.[24,25] The exponential temperature dependence of R_{IHD} in Eq. (11) is due to the Arrhenius-type dependence of τ_c (σ and $|\sigma|/3$ are the dimensionless barrier heights for the cubic anisotropy potential[17]). We remark that our results pertain to the memoryless (white noise) limit (Ohmic damping). However, as conjectured in Ref. 23, they should also hold for long-time memory, with a reduced effective dissipation constant.

Expressions (10) and (11) give an accurate simplified description of SR for cubic anisotropy in the low-temperature limit and are compared with the exact matrix continued fraction solution[17,18] in Figs. 2 and 3. A striking feature of magnetic SR for cubic anisotropy is that the SNR curve intrinsically depends on the damping parameter α. This behavior is an explicit signature of *precession aided relaxation*; i.e., the coupling between the relaxational and precessional magnetodynamic modes arising from the nonaxially symmetric potential. Here, at each temperature, there exists a certain α_{max} for which the SNR has a maximum R_{max}. The analytic form of Eqs. (11) and (10) allows one to estimate readily the peak magnitude R_{max} and the half-width δR of the damping dependence of R, $R_{\mathrm{max}} \approx R_{\mathrm{IHD}}|_{\alpha\to 0}$ (see Fig. 2) and δR (which can be evaluated from the solution of the algebraic equation $R_{\mathrm{IHD}} = R_{\mathrm{max}}/2$). These estimations yield $R_{\mathrm{max}} \sim 8\sigma^2 e^{-\sigma}/\pi$, $\delta R \sim (\sqrt{14}+\sqrt{2})/2$ for $K_c > 0$ and $R_{\mathrm{max}} \sim 8\sigma^2 e^{-|\sigma|/3}/(3\pi)$, $\delta R \sim (\sqrt{14}-\sqrt{2})/2$ for $K_c < 0$, respectively.

The strong magnetic mode coupling exhibited by cubic crystals suggests a way of determining the damping coeffi-

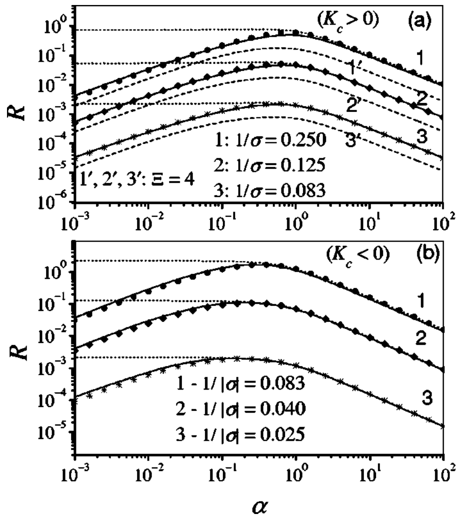

FIG. 2. SNR vs α for various values of $1/\sigma$. Solid lines, Eq. (7), exact matrix continued fraction solution (Refs. 17 and 18). Symbols, calculations from Eq. (10). Dotted lines, the SNR in the IHD limit given by Eq. (11). Dashed lines, the SNR for cubic+uniaxial anisotropy with $\Xi = 4$ [here the factor $3\langle\cos^2\vartheta(0)\rangle_0$ must be added into Eq. (7).

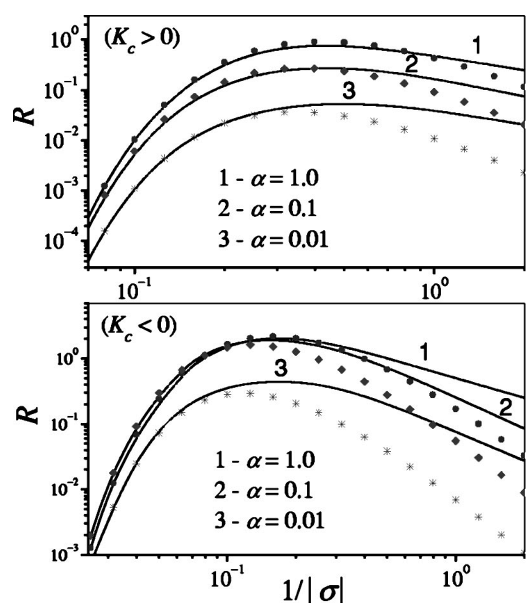

FIG. 3. SNR vs $1/\sigma$ for $\alpha = 1.0$ (1), 0.1 (2), 0.01 (3). Solid lines, Eq. (7), exact matrix continued fraction solution (Refs. 17 and 18). Symbols, calculations from Eq. (10).

cient from measurements of SNR in a fine-particle system. For example, one may fit the theory to the experimental SNR, Eq. (4), at a given frequency Ω, so that the only fitting parameter is α. This procedure will allow one to estimate the damping parameter (e.g., by adjusting δR and $\alpha_{\max}$). Moreover, α can also be determined for different T, so yielding its temperature dependence. Such experiments are much more easily accomplished than those for uniaxial particles in the presence of a symmetry breaking strong dc external magnetic field.[13,26,27] These considerations suggest that the present problem is of interest in both its fundamental and technical physics aspects. From a fundamental viewpoint a knowledge of $\alpha(T)$ is required for studies of various possible mechanisms of orientational reversal of **M**, e.g., macroscopic quantum tunneling,[28] as $\alpha(T)$ allows one to distinguish and separate the various relaxation mechanisms. In applied physics, the dependence $\alpha(T)$ is essential for the development of the various techniques generically known as thermal-assisted magnetic recording on single grains.

Other types of nonaxially symmetric anisotropies can be treated in a similar manner. For illustration, we present in Fig. 2(a) the calculations of the SNR for mixed cubic and uniaxial anisotropies evaluated for different values of $h = \Xi/\sigma = 1.0$, 0.5, and 0.33 and for $K_c > 0$. Here the effect of the uniaxial anisotropy is of importance as the parameter h now determines the barrier heights of the (cubic+unaxial) potential and yields the contribution to the decrease of the SNR, $\sim e^{-\sigma(1+h)}$. A detailed treatment of the SNR in the cubic+unaxial potential will be given elsewhere.

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