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2010 J. Phys.: Condens. Matter 22 376001

(http://iopscience.iop.org/0953-8984/22/37/376001)

IOP Publishing | Journal of Physics: Condensed Matter
J. Phys.: Condens. Matter **22** (2010) 376001 (7pp) doi:10.1088/0953-8984/22/37/376001

Classical–quantum crossover in magnetic stochastic resonance in uniaxial superparamagnets

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Received 20 May 2010, in final form 2 August 2010
Published 31 August 2010
Online at stacks.iop.org/JPhysCM/22/376001

Abstract
It is shown that the signal-to-noise ratio of the magnetic moment fluctuations in the magnetic stochastic resonance of a quantum uniaxial paramagnet of arbitrary spin value S subjected to a weak probing ac field $\mathbf{H}(t) = \mathbf{H}\cos\Omega t$ and a dc bias magnetic field $\mathbf{H}_0$ displays a pronounced dependence on S. The dependence arises from the quantum dynamics of spins which differs markedly from the magnetization dynamics of classical superparamagnets. In the large spin limit, $S \to \infty$, the quantum solutions reduce to those for a classical uniaxial superparamagnet.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

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 [1–3]. The archetypal theoretical model of SR [1] is a 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 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-noise 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.

The behavior of magnetic nanosystems (such as superparamagnetic particles, nanoclusters, and molecular magnets) forced by a weak ac magnetic field is yet another important manifestation of SR. Here the magnetic anisotropy provides the multistable states for the magnetization $\mathbf{M}$ while the thermal fluctuations or random field 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 again may be defined as the enhancement of the SNR due to noise [4]. The magnetic SR has been predicted first theoretically [5–7] and shortly afterward observed experimentally [8]. The SNR of the magnetic moment fluctuations is of some interest because nanoparticle magnetism is a rapidly expanding area of research with many novel applications. These arise both in the (applied) area of information storage and in other (fundamental) aspects such as the crossover between classical and quantum behavior of the magnetization since single-domain particles exhibit essentially classical behavior while smaller entities such as free nanoclusters made of many atoms, molecular clusters, and molecular magnets exhibit pronounced quantum behavior.

The main features of the magnetic SR in single-domain particles [9–13] may be completely understood in terms of the classical (macrospin) model of the coherent rotation of the magnetization [14]. Here each particle behaves like a paramagnetic atom having a magnetic moment $\sim 10^4$–10^5 Bohr magnetons, i.e., $S \sim 10^4$–10^5. In the presence of a dc bias field, the normalized magnetic free energy density V of a *uniaxial* single-domain particle is given by the asymmetric bistable potential

$$V(\vartheta) = -kTv^{-1}\sigma(\cos^2\vartheta + 2h\cos\vartheta), \tag{1}$$

where ϑ is the polar angle, $\sigma = vK'/(kT)$ is the dimensionless barrier height parameter, v is the volume of the particle, k is Boltzmann's constant, T is the absolute temperature, K' is the anisotropy constant, and $h = M_s H_0/(2K')$ is the bias field parameter (M_s is the saturation magnetization). In the absence of the dc field, the magnetization of the uniaxial particle has two equivalent stable orientations at $\vartheta = 0$ and π, so that it is an ideal example of a bistable system subjected to noise. Here the reversal of the classical spin is due to thermal activation and the rate of transitions between the potential wells is controlled by the parameter σ, which relates the height $K'v$ of the magnetic anisotropy barrier to the thermal energy. Thus one may regard the inverse of σ as the dimensionless temperature, i.e., the noise intensity. A dc bias field $\mathbf{H}_0$ applied to the particle parallel to its anisotropy axis breaks the bidirectional symmetry of the potential. However, an asymmetric two-minima profile of the potential $V(\vartheta)$ survives as long as the bias field parameter $h = M_s H_0/(2K') \leqslant 1$.

In contrast, we have little knowledge about magnetic SR in superparamagnets with *smaller* spin values $S \sim 10$–100, where quantum effects and quantum-classical crossover appear. Here the spin reversal is either due to thermal activation or tunneling or a combination of both and quantum effects appear. These quantum effects are not the same as those in the SR for translational Brownian motion (see, e.g., [15, 16] and references cited therein) because in spite of some analogies the quantum spin dynamics essentially differ from those of Brownian particles owing to the different symmetries of the groups of rotations and translations. Here we shall treat quantum effects in the SR of the magnetic moment fluctuations for spin systems taking as an example a uniaxial paramagnet of *arbitrary* spin value S in an external dc magnetic field $\mathbf{H}_0$ and a weak probing ac field $\mathbf{H}(t) = \mathbf{H}\cos\Omega t$ applied along the Z axis, i.e., the axis of symmetry. Thus the system Hamiltonian has the form

$$\hat{H} = \hat{H}_S + \hat{H}_{SB} + \hat{H}_B,$$

where

$$\hat{H}_S = -KS^{-2}\hat{S}_Z^2 - \gamma\hbar[H_0 + H\cos(\Omega t)]\hat{S}_Z \tag{2}$$

$\hat{S}_Z$ is the Z-component of the spin operator $\mathbf{S}$, $\hbar$ is Planck's constant, and γ is the gyromagnetic ratio, $\hat{H}_{SB}$ describes interaction of the spin with the thermostat, and $\hat{H}_B$ characterizes the thermostat. This Hamiltonian includes a uniaxial anisotropy term plus the Zeeman coupling with the external field, comprising a generic model for quantum relaxation phenomena in uniaxial spin systems such as molecular magnets, nanoclusters, etc (see, e.g., [17, 18] and references cited therein). Now Garanin [17] and also García-Palacios and Zueco [18] have recently considered the longitudinal relaxation of quantum superparamagnets with Hamiltonian (2) for arbitrary S using the spin density matrix in the second order of perturbation theory in the spin-bath coupling. They gave a concise treatment of the spin dynamics using the quantum Hubbard operator representation of the evolution equation for the spin density matrix. The nonlinear relaxation of uniaxial quantum spin systems has also been treated by Kalmykov *et al* [19] via the respective evolution equations for the reduced density matrix and corresponding phase space distribution function in the high temperature, Ohmic damping, and weak spin-bath coupling limits using the methods already available for classical spins. In the large spin limit, their quantum solutions reduce to those yielded by the Fokker–Planck equation for a classical uniaxial superparamagnet [13, 20–24] while for linear response, the results entirely agree with those given in [17, 18].

2. Basic equations for SNR of uniaxial paramagnets

Here we shall treat the spin size effects in the magnetic SR for uniaxial quantum paramagnets using the above spin-boson model via Kubo's linear response theory [25]. A typical Fourier component M_ω of the longitudinal components of the magnetization of a spin system is related to the corresponding Fourier component of a weak applied ac field H_ω through the complex magnetic susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, namely,

$$M_\omega = \chi(\omega)H_\omega. \tag{3}$$

According to linear response theory [25], $\chi(\omega)$ is defined as

$$\frac{\chi(\omega)}{\chi_0} = 1 - i\omega\int_0^\infty C(t)e^{-i\omega t}\,dt, \tag{4}$$

where

$$C(t) = \frac{\left\langle \int_0^\beta [\hat{S}_Z(-i\lambda\hbar) - \langle\hat{S}_Z\rangle_0][\hat{S}_Z(t) - \langle\hat{S}_Z\rangle_0]\,d\lambda \right\rangle_0}{\left\langle \int_0^\beta [\hat{S}_Z(-i\lambda\hbar) - \langle\hat{S}_Z\rangle_0][\hat{S}_Z(0) - \langle\hat{S}_Z\rangle_0]\,d\lambda \right\rangle_0} \tag{5}$$

is the normalized equilibrium correlation function,

$$\chi_0 = (\gamma\hbar)^2 \left\langle \int_0^\beta [\hat{S}_Z(-i\lambda\hbar) - \langle\hat{S}_Z\rangle_0][\hat{S}_Z(0) - \langle\hat{S}_Z\rangle_0]\,d\lambda \right\rangle_0 \tag{6}$$

is the static susceptibility, the brackets $\langle\,\rangle_0$ denote the equilibrium statistical average, and $\beta = (kT)^{-1}$. 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 $\Phi_M^{(s)}(\Omega) = (H|\chi(\Omega)|)^2/2$ while the noise-induced part $\Phi_M^{(n)}(\Omega) = \chi''(\Omega)/(\pi\beta\Omega)$ is obtained using the fluctuation-dissipation theorem [1, 13]. Thus on combining the above equations, one obtains the SNR $= \Phi_M^{(s)}/\Phi_M^{(n)}$ of the magnetic moment fluctuations as

$$\mathrm{SNR} = \frac{\pi\Omega\beta H^2|\chi(\Omega)|^2}{2\chi''(\Omega)}. \tag{7}$$

The linear response theory result equation (7) is very useful because of its generality. It shows that the calculation of the SNR may be reduced to that of the dynamic susceptibility $\chi(\Omega)$, which is a fundamental dynamical characteristic of any relaxing system. For uniaxial paramagnets with Hamiltonian (2), $\chi(\Omega)$ has been calculated recently in [18, 19]. Using these results, we now estimate quantum effects in the magnetic SR.

According to equation (4), the behavior of $\chi(\Omega)$ in the frequency domain is completely determined by the time behavior of the correlation function $C(t)$. For uniaxial paramagnets, $C(t)$ may be written as the *finite discrete* set of relaxation modes [18, 19]

$$C(t) = \sum_{k=1}^{2S} c_k \mathrm{e}^{-\lambda_k t}, \tag{8}$$

where λ_k are the eigenvalues of the (finite, in the quantum case) system matrix and $\sum_{k=1}^{2S} c_k = 1$. Consequently, equations (4) and (8) allow us to write $\chi(\Omega)$ as the finite sum of Lorentzians [18, 19]

$$\frac{\chi(\Omega)}{\chi_0} = \sum_{k=1}^{2S} \frac{c_k}{1 + \mathrm{i}\Omega/\lambda_k}. \tag{9}$$

Consequently, the asymptotic behavior of $\chi(\Omega)$ in the extremes of very low and very high frequencies is given by

$$\frac{\chi(\Omega)}{\chi_0} \sim \begin{cases} 1 - \mathrm{i}\Omega\tau_{\mathrm{cor}} + \cdots, & \Omega \to 0, \\ -\mathrm{i}(\Omega\tau_{\mathrm{ef}})^{-1} + \cdots, & \Omega \to \infty, \end{cases} \tag{10}$$

where

$$\tau_{\mathrm{cor}} = \sum_{k=1}^{2S} c_k/\lambda_k \qquad \text{and} \qquad \tau_{\mathrm{ef}} = \left(\sum_{k=1}^{2S} \lambda_k c_k\right)^{-1}. \tag{11}$$

We remark that the relaxation times so defined τ_{cor} and τ_{ef} parameterize the time behavior of the correlation function $C(t)$. Indeed, the integral relaxation time τ_{cor} is the area under $C(t)$, namely, $\tau_{\mathrm{cor}} = \int_0^\infty C(t)\,\mathrm{d}t$, and the effective relaxation time $\tau_{\mathrm{ef}} = -1/\dot{C}(0)$ gives precise information on the initial decay of $C(t)$ in the time domain. The relaxation times τ_{cor} and τ_{ef} can equivalently be given by the analytic formulas [17–19]

$$\tau_{\mathrm{ef}} = \frac{2\chi_S \tau_N}{\sum_{k=1-S}^{S} [S(S+1) - k(k-1)]\rho_k}, \tag{12}$$

$$\tau_{\mathrm{cor}} = \frac{2\tau_N}{\chi_S} \sum_{k=1-S}^{S} \frac{\left[\sum_{m=k}^{S} (m - \langle \hat{S}_Z \rangle_0)\rho_m\right]^2}{[S(S+1) - k(k-1)]\rho_k}, \tag{13}$$

where $\chi_S = \sum_{m=-S}^{S} m^2 \rho_m - (\sum_{m=-S}^{S} m\rho_m)^2$, $\langle \hat{S}_Z \rangle_0 = \sum_{m=-S}^{S} m\rho_m$, the $\rho_m = \mathrm{e}^{\frac{\sigma}{S^2}m^2 + \frac{2\sigma h}{S}m}/Z_S$ are the matrix elements of the equilibrium density matrix, $Z_S = \sum_{m=-S}^{S} \mathrm{e}^{\frac{\sigma}{S^2}m^2 + \frac{2\sigma h}{S}m}$ is the partition function, $\sigma = \beta K$, $h = \hbar\gamma S H_0/(2K)$ is a reduced field parameter, and τ_N is a characteristic free diffusion time.

Equations (12) and (13) have been derived in [19] in the weak coupling limit and for Ohmic damping. Thus, the correlation time characterizing the thermal bath is short enough to allow one to approximate the stochastic process originating in it by a Markov process. These approximations may be used in the high temperature limit, $\beta|\varepsilon_m - \varepsilon_{m\pm1}| \ll 1$, where $\varepsilon_m, \varepsilon_{m\pm1}$ are eigenvalues of the energy. In the parameter range, where they fail (e.g., throughout the very low temperature region), more general forms of the phase space and density matrix equations must be used (given, e.g., in [17, 18]). Nevertheless, we shall still use the model based on the above approximation because despite many drawbacks it qualitatively describes the relaxation in spin systems. Moreover, the model can be regarded as the direct quantum generalization of the Langevin formalism used by Brown [14] in his theory of relaxation of classical superparamagnetic particles.

By analogy with the SNR of the magnetic moment fluctuations for a classical superparamagnet [13], equation (7) can be presented as $\mathrm{SNR} = \pi(\beta\gamma\hbar SH)^2 R_\Omega/(2\tau_N\sigma)$, where the dimensionless SNR factor R_Ω is given by

$$R_\Omega = \frac{\sigma\tau_N\Omega|\chi(\Omega)|^2}{\beta(\gamma\hbar S)^2\chi''(\Omega)}. \tag{14}$$

Thus the relevant quantity is R_Ω. In general, R_Ω, besides the obvious dependence as in the classical case on the noise intensity (temperature), the constant (bias) field strength h, and the frequency of the exciting field Ω, also depends on the spin value S. In the adiabatic, $\Omega \to 0$, and very high-frequency, $\Omega \to \infty$, limits, equation (14) can be considerably simplified yielding

$$R_0 = \tau_N\sigma\chi_S/(\tau_{\mathrm{cor}}S^2) \tag{15}$$

and

$$R_\infty = \tau_N\sigma\chi_S/(\tau_{\mathrm{ef}}S^2). \tag{16}$$

In the classical limit, $S \to \infty$, the above results agree in all respects with the corresponding classical solutions [13] (see appendix).

3. Results and discussion

As shown in [18, 19], two distinct bands appear in the spectrum of the imaginary part $\chi''(\Omega)$ of the susceptibility for a uniaxial quantum paramagnet. The low-frequency band is due to the slowest ('interwell') relaxation mode. The characteristic frequency and the half-width of this band are determined by the smallest nonvanishing eigenvalue λ_1 [18, 19]. We remark that the smallest eigenvalue λ_1 is associated with the long time behavior of $C(t) \sim \mathrm{e}^{-t/\tau}$, $t \gg \tau$, which is dominated by the longest relaxation (or the reversal) time of the magnetization τ. The high-frequency band in $\chi''(\Omega)$ is due to the individual near degenerate high-frequency modes corresponding to the eigenvalues $\lambda_k \gg \lambda_1$ ($2S \geqslant k \geqslant 2$). Thus, if one is interested solely in the low-frequency region ($\Omega\tau \leqslant 1$), where their effect may be ignored, the dynamic susceptibility $\chi(\Omega)$ may be approximated as the single Lorentzian [19]

$$\frac{\chi(\Omega)}{\chi_0} \approx 1 - \frac{\mathrm{i}\Omega\tau_{\mathrm{cor}}}{1 + \mathrm{i}\Omega/\lambda_1}, \tag{17}$$

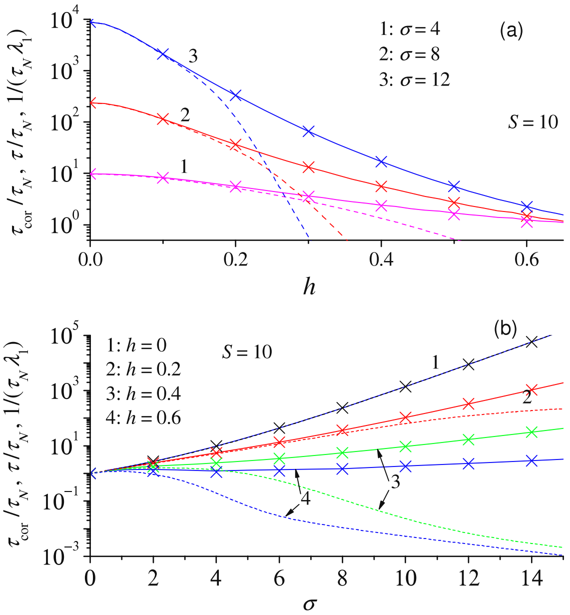

Figure 1. Integral relaxation time τ_{cor}, equation (13) (dashed lines), λ_1^{-1} (crosses), and its analytic approximation τ as rendered by equation (18) (solid lines) as functions of (a) the field parameter h for various values of the anisotropy (inverse temperature) parameter σ and vice versa as functions of (b) the anisotropy (inverse temperature) parameter σ for various values of the field parameter h both for $S = 10$.

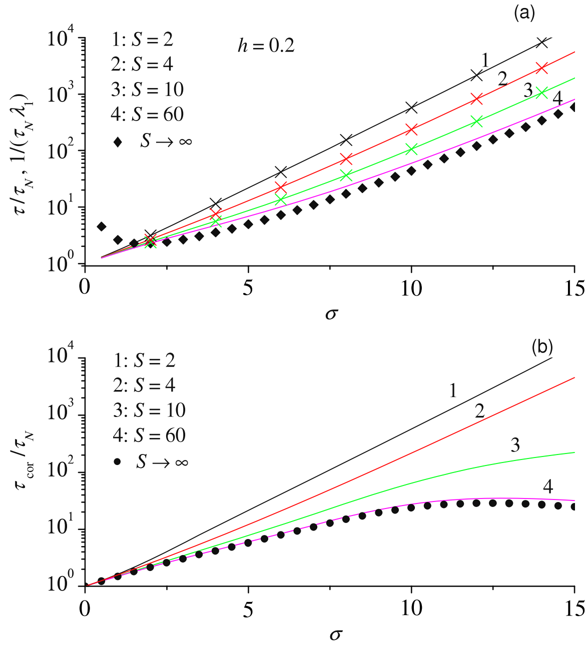

Figure 2. Integral relaxation time τ_{cor}, equation (13), λ_1^{-1} (crosses), and its analytic approximation τ, equation (18), as functions of σ for $h = 0.2$ and various S. Diamonds and filled circles: classical limit, $S \to \infty$, equations (A.6) and (A.4), respectively.

where λ_1^{-1} can also be approximately evaluated as [18]

$$\lambda_1^{-1} = \frac{2\tau_N}{\chi_\Delta} \times \sum_{k=1-S}^{S} \frac{\sum_{n=k}^{S} \rho_n (n - \langle \hat{S}_Z \rangle_0) \sum_{m=-S}^{k-1} [\Delta - \mathrm{sgn}(m - m_b)]\rho_m}{[S(S+1) - k(k-1)]\rho_k}. \quad (18)$$

Here $\Delta = \sum_{m=-S}^{S} \mathrm{sgn}(m - m_b)\rho_m$ and

$$\chi_\Delta = \sum_{m=-S}^{S} m\, \mathrm{sgn}(m - m_b)\rho_m - \left(\sum_{m=-S}^{S} m\rho_m\right) \times \left(\sum_{m=-S}^{S} \mathrm{sgn}(m - m_b)\rho_m\right),$$

where

$$m_b = -(S - 1/2)h - 1/2 + \mathrm{Fp}[(S - 1/2)h + S + 1/2]$$

is the quantum number corresponding to the top of the barrier and Fp[a] denotes the fractional part of a. In the absence of a dc field ($h = 0$), equation (18) reduces to a closed analytical expression for τ given by Villain *et al* and Würger [26].

Equations (14) and (17) indicate that the low-frequency behavior of the SNR of a quantum paramagnet is mainly determined by τ_{cor} and τ. The dependences of τ_{cor} and τ on the model parameters (external field, anisotropy constants, and spin value) may, however, differ markedly. For example, the behavior of τ_{cor} and τ is similar for small dc external fields only. In strong dc fields, τ can *diverge exponentially* from τ_{cor} as in a classical uniaxial paramagnet. This effect was discovered numerically by Coffey *et al* [20] for classical superparamagnets and later explained quantitatively by Garanin [21] (see also [13], chapter 1 for details). He showed analytically that the contribution of relaxation modes other than the overbarrier one to the overall relaxation process becomes significant for high external fields due to population depletion of the shallower of the two potential wells of a bistable potential under the action of a strong external dc field. The effect being manifested in the exponential divergence of the two relaxation times. Comparison of τ_{cor} and τ as functions of the field parameter h for various values of σ and as a function of σ for various values of h are given in figure 1. The times τ_{cor} and τ as functions of the anisotropy parameter σ for various values of the spin value S are shown in figure 2. Here τ has been evaluated both by using the *approximate* equation (18) and by calculating *numerically* the smallest eigenvalue of the system matrix (as described in [19]). For values of the field $h < 0.5$, the relative deviation of τ calculated by either method does not exceed 1%. We see that the figures 1 and 2 display a pronounced dependence of both τ_{cor} and τ on the field, anisotropy, and spin parameters. It is apparent from figure 2 that for large S, the quantum solutions reduce to the corresponding classical ones, however, they can deviate strongly from each other for small S. Typical values of S for the quantum-classical crossover are ∼20–50. In general, the smaller the anisotropy σ is the smaller the S number required for convergence of the quantum equations to the classical ones [19].

The SNR in the adiabatic limit $\Omega = 0$, $R_{\Omega=0}$, versus the dimensionless temperature parameter σ^{-1} is shown in figures 3 and 4 for various values of S and h. The maximum in R_0 is

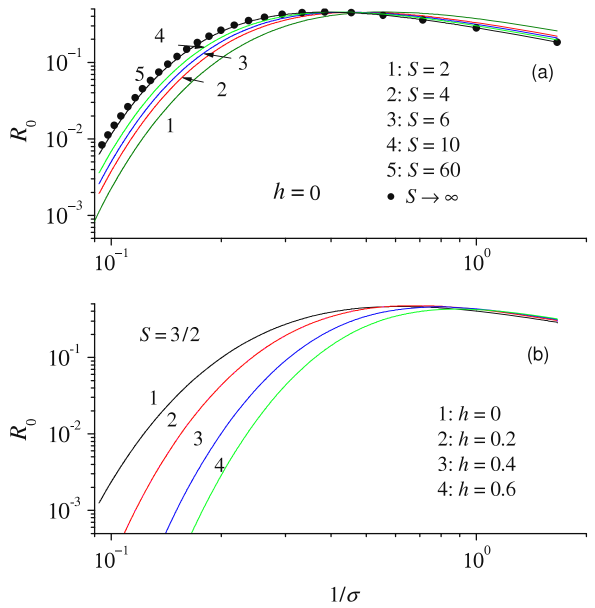

Figure 3. SNR in the adiabatic limit $\Omega = 0$ as a function of dimensionless temperature σ^{-1} (a) for various S and $h = 0$ and (b) for various values of the field parameter h and $S = 3/2$. Solid lines: equation (15). Filled circles: classical limit, $S \to \infty$, equations (A.2) and (A.4).

attained at $\sigma_{\max}^{-1} \sim 0.4$–$0.6$ and it shifts to low temperatures with increasing S (for the molecular magnet Mn_{12} acetate with $S = 10$, $\sigma_{\max}^{-1} \approx 0.45$ corresponds to $T \sim 30$ K). Moreover, the maximum of the SNR moves to higher temperatures with increasing h. In the limit $\sigma^{-1} \to 0$, $R_0 \to 0$ for small S and h while S increases at finite h, $R_0 \to$ constant (see figure 4) because of the temperature dependence of τ_{cor} (see figures 1 and 2) which on increasing S and h, τ_{cor} progressively loses its Arrhenius character. The explanation for this shift follows because the bias field radically alters the temperature dependence of the static susceptibility χ_S of the system [27]. In a nonzero bias field, the effect of saturation of the longitudinal magnetization is crucial causing χ_S to tend to zero at zero temperature. In general, we see that the quantum effects can lead to both *amplification* and *attenuation* of the SNR. Meanwhile, we recall that in the classical limit, $S \to \infty$, and, λ_1 from equation (A.6) is exponentially small for $\sigma \gg 1$ and decreases rapidly as the system is cooled, while all the other eigenvalues of the system matrix λ_k have a non-exponential dependence on σ. Hence, at any finite frequency Ω (i.e., outside the adiabatic limit), the ratio Ω/λ_1 tends to infinity with decreasing temperature, $T \to 0$, even at very low frequencies since the *interwell* transition is almost 'frozen out'. In spite of this, the spin, although confined to a particular potential well, is *not yet completely immobilized* and can still take part in *intrawell* motion. Thus for $\Omega \neq 0$, $R_\Omega \to$ const as $\sigma \gg 1$ (see figure 5). The function R_Ω versus the dimensionless frequency $\Omega\tau_N$ is presented in figures 6 and 7 exemplifying the quantum effects. Here the SNR is a monotonically increasing function from the low-frequency limit R_0 right through to its plateau value R_∞ given by equation (16).

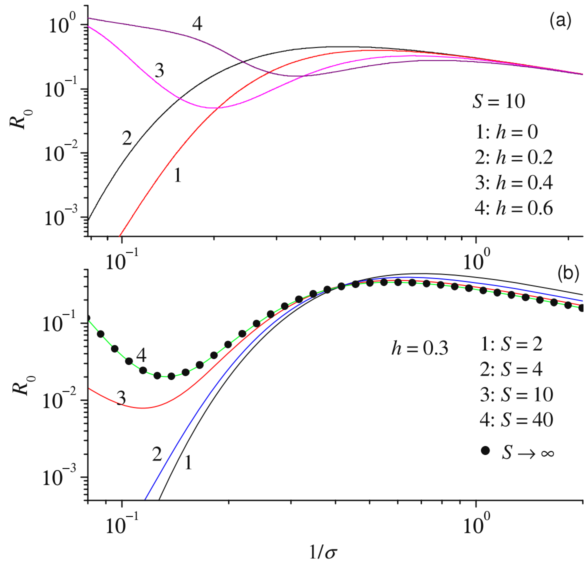

Figure 4. SNR in the adiabatic limit $\Omega = 0$ as a function of σ^{-1} (a) for various values of h and $S = 10$ and (b) for various values of S and $h = 0.3$. Solid lines: equation (15). Filled circles: classical limit $S \to \infty$, equations (A.2) and (A.4).

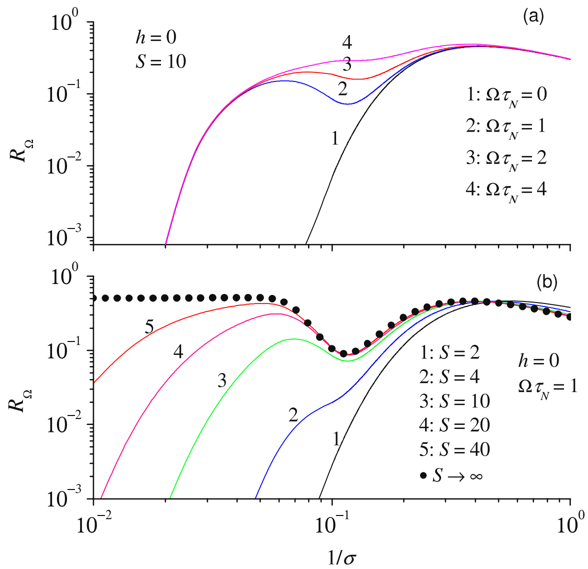

Figure 5. SNR as a function of σ^{-1} (a) for various values of $\Omega\tau_N$ and $S = 10$ and (b) for various values of S and $\Omega\tau_N = 1$ in the absence of the dc bias field ($h = 0$). Solid lines: the quantum equations (9) and (14). Filled circles: classical limit, $S \to \infty$.

4. Concluding remarks

We have studied the magnetic SR of a quantum uniaxial superparamagnet of arbitrary spin S in the high temperature and weak spin-bath coupling limit. The principal result is that one may determine the transition from the SR corresponding to quantum elementary spin relaxation to that pertaining to a giant spin as a function of the spin size S. In other words, one can study the evolution of the SR from that an elementary spin to molecular magnets ($S \sim 10$) to nanoclusters ($S \sim 100$), and to classical superparamagnetic particles ($S \geqslant 1000$). Hence, one

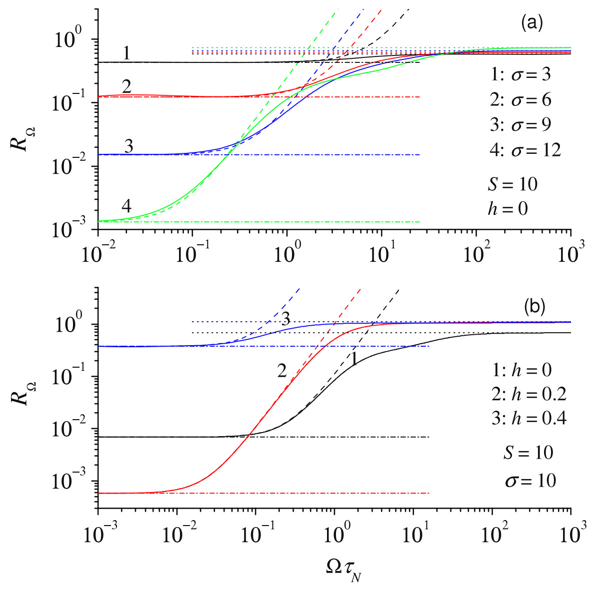

Figure 6. SNR as a function of $\Omega\tau_N$ (a) for various values of σ, $S = 10$, and $h = 0$ and (b) for various values of h and $S = 10$, $\sigma = 10$. Solid lines: equations (9) and (14). Dashed lines: the low-frequency quantum equations (17) and (14). Dashed–dotted and dotted lines: the adiabatic, equation (15), and high-frequency, equation (16), limits, respectively.

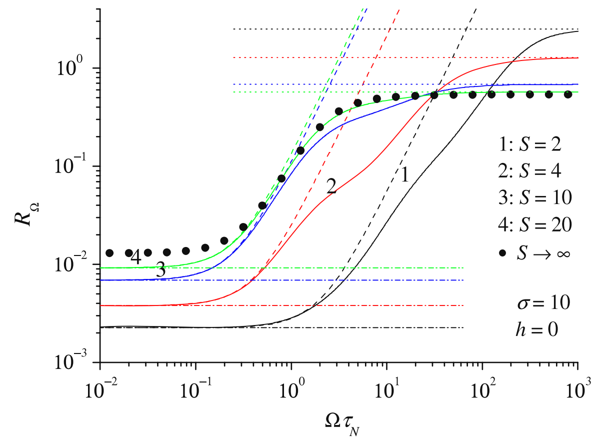

Figure 7. SNR as a function of $\Omega\tau_N$ for various values of S and $\sigma = 10$, $h = 0$. Solid lines: equations (9) and (14). Dashed lines: the low-frequency equations (17) and (14). Dashed–dotted and dotted lines: the adiabatic, equation (15), and high-frequency, equation (16), limits, respectively. Filled circles: classical limit, $S \to \infty$.

may accurately estimate the value of S (typically in the range 20–50) at which the crossover to classical superparamagnetic behavior takes place [28]. Thus one may assign a range of validity as a function of the spin size to the classical Néel–Brown treatment of a superparamagnetic particle with the simplest uniaxial anisotropy and Zeeman energy.

Here we have considered the SR in uniaxial superparamagnets in weak ac fields and in the simplest configuration (the direction of the ac and dc magnetic fields coincides with the easy axis of the magnetization). However, the method may be generalized to other interesting cases such as the nonlinear SR in strong ac fields and arbitrary directions of applied fields. Furthermore, the relatively elementary calculation outlined above is of particular interest as a basis for future understanding of the SR of spin systems characterized by nonaxially symmetric Hamiltonians commonly used to describe the magnetic properties of molecular magnets and nanoclusters. Now as shown for classical superparamagnets [12], the SNR in the magnetic stochastic resonance of single-domain ferromagnetic nanoparticles having *nonaxially symmetric* magnetocrystalline 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. The effect, which does not exist for axially symmetric potentials, may be used to determine α for quantum spin systems by means of the SR effect. The extension to particular nonaxially symmetric spin systems such as biaxial, cubic, etc would also allow one to include spin size effects in important technological applications involving magnetic relaxation where tunneling in the presence of a transverse field influences the behavior of the reversal time, the switching and hysteresis curves, etc. Furthermore, our approach can also be applied to the calculation of the nonlinear effects in the magnetic SR of quantum superparamagnets driven by a strong ac magnetic field by generalizing the matrix continued fraction method of solution of the Fokker–Planck equation for the ac nonlinear response of classical spins [29].

Acknowledgment

This publication has emanated from research conducted with the financial support of FP7-PEOPLE-Marie Curie Actions (Project No. 230785 NANOMAGNETS).

Appendix. Classical limit

In the classical limit, $S \to \infty$, the normalized longitudinal dynamic susceptibility $\chi(\Omega)$ is also given by equation (4), where now $C(t)$ from (5) becomes

$$C(t) = \frac{\langle \cos\vartheta(0)\cos\vartheta(t)\rangle_0 - \langle\cos\vartheta\rangle_0^2}{\langle\cos^2\vartheta\rangle_0 - \langle\cos\vartheta\rangle_0^2}. \tag{A.1}$$

Here $\langle\cos\vartheta\rangle_0$ and $\langle\cos^2\vartheta\rangle_0$ can be calculated analytically yielding [13]

$$\langle\cos\vartheta\rangle_0 = \frac{1}{Z}\int_{-1}^{1} x\mathrm{e}^{-\beta V(x)}\,\mathrm{d}x = \frac{\mathrm{e}^{\sigma}\sinh(2\sigma h)}{\sigma Z} - h,$$

$$\langle\cos^2\vartheta\rangle_0 = \frac{1}{Z}\int_{-1}^{1} x^2\mathrm{e}^{-\beta V(x)}\,\mathrm{d}x = \frac{\mathrm{e}^{\sigma}[\cosh(2\sigma h) - h\sinh(2\sigma h)]}{\sigma Z} + h^2 - \frac{1}{2\sigma},$$

where $\beta V(x) = -\sigma(x^2 + 2hx)$,

$$Z = \int_{-1}^{1}\mathrm{e}^{-\beta V(x)}\,\mathrm{d}x = \frac{1}{2}\sqrt{\frac{\pi}{\sigma}}\mathrm{e}^{-\sigma h^2}\{\mathrm{erfi}[(1+h)\sqrt{\sigma}] + \mathrm{erfi}[(1-h)\sqrt{\sigma}]\}$$

is the partition function, and $\mathrm{erfi}(x) = (2/\sqrt{\pi})\int_0^x e^{t^2}\,dt$ is the error function of imaginary argument. A simple analytic equation for $\chi(\Omega)$ for a classical superparamagnet is given in [21, 24].

The classical analogues of equations (15) and (16) are

$$R_0 = \frac{\tau_N\sigma}{\tau_{\mathrm{cor}}}(\langle\cos^2\vartheta\rangle_0 - \langle\cos\vartheta\rangle_0^2), \qquad (A.2)$$

$$R_\infty = \frac{\tau_N\sigma}{\tau_{\mathrm{ef}}}(\langle\cos^2\vartheta\rangle_0 - \langle\cos\vartheta\rangle_0^2). \qquad (A.3)$$

τ_{cor} and τ_{ef} can now be expressed instead of summations in closed integral form as [13, 24]

$$\tau_{\mathrm{cor}} = \frac{2\tau_N}{Z(\langle\cos^2\vartheta\rangle_0 - \langle\cos\vartheta\rangle_0^2)} \times \int_{-1}^{1}\left[\int_{-1}^{x}(z - \langle\cos\vartheta\rangle_0)e^{-\beta V(z)}\,dz\right]^2 \frac{e^{\beta V(x)}}{1-x^2}\,dx, \qquad (A.4)$$

$$\tau_{\mathrm{ef}} = 2\tau_N\frac{\langle\cos^2\vartheta\rangle_0 - \langle\cos\vartheta\rangle_0^2}{1 - \langle\cos^2\vartheta\rangle_0}. \qquad (A.5)$$

Finally, at low temperatures, $\sigma \gg 1$, the smallest nonvanishing eigenvalue $\lambda_1 = \tau^{-1}$ can be closely approximated by Brown's asymptotic high energy barrier formula for the magnetization reversal time of a uniaxial superparamagnetic particle in the presence of a dc field, namely [14],

$$\lambda_1 = \tau^{-1} \approx e^{-\sigma(1-h)^2}\sigma^{3/2}\frac{(1-h^2)}{\tau_N\sqrt{\pi}}[1 - h + (1+h)e^{-4\sigma h}]. \qquad (A.6)$$

For $h = 0$, the corresponding equations for λ_1, τ_{cor}, and τ_{ef} are given in [13, 30].

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