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RELAXATION OF THE DENSITY MATRIX FOR AN ARBITRARY SPIN IN RANDOM EXTERNAL FIELDS

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НОВОСИБИРСК

RELAXATION OF THE DENSITY MATRIX FOR AN ARBITRARY SPIN IN RANDOM EXTERNAL FIELDS

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Abstract

The relaxation of the density matrix for an arbitrary spin S under magnetic noise is independent of S under the assumption that the g-factor is S-independent. However, the decrements of polarization moments increase with the multipolarity l as $l(l+1)$. For isotropic noise created by external fields of arbitrary multipolarities the relaxation is also S-independent in the classical limit $S \to \infty$ under the analogous assumption. In the case of anisotropic noise of the multipolarity higher than unity the relaxation of a pure state is described in the limit $S \gg l \gg 1$ by the heat equation. Here under the same assumption the relaxation falls off at $S \to \infty$ as $1/S^2$. If no such assumption on the S-dependence of the interaction parameters is made, the relaxation under magnetic noise and arbitrary isotropic one may depend on S, but this dependence remains smooth in the limit $S \to \infty$. The strong-noise situation is also considered when the characteristic phase shift at a kick is not small and one cannot restrict to pair correlators.

1 Introduction

The problem of the relaxation of the spin density matrix in random external fields is of great applied interest, in particular for magnetic resonance and atomic spectroscopy. In the present article more theoretical aspects of the problem will be studied: how the relaxation of polarization moments depends on their multipolarity l, as well as on the spin magnitude S, in particular at the transition to the classical limit $S \to \infty$.

For some special cases the result for the last problem is fairly obvious. The motion equations for the polarization vector in arbitrary magnetic fields are the same at any S as long as the g-factor is S-independent. Therefore, the relaxation of this vector in random magnetic fields is also S-independent. The same result holds for the relaxation of any polarization moment under magnetic noise. On the other hand, the decrements of polarization moments grow with their multipolarities l as $l(l+1)$.

The next simple case is the density matrix relaxation under a spatially isotropic noise of an arbitrary multipolarity. It is quite evident that under isotropic noise the density matrix parameters of any multipolarity relax independently. As to the decrement behaviour with S, it depends on the assumptions made about the dependence on S of the interaction parameters. If they have finite limits at $S \to \infty$ which is a natural generalization of the assumption made in the magnetic case, the decrements are finite as well. Anyway, those decrements depend on S smoothly, even for $S \gg 1$.

The conclusions made are not specific to the case of "weak" noise when the phase shift under each kick is small and one can restrict to pair correlators. They are demonstrated to be valid in the case of strong noise as well.

It is only natural to go over to a spatially anisotropic noise of higher multipolarity, than magnetic one. Here the relaxation of the density matrix corresponding to a pure state is described in the limit $S \gg l \gg 1$ by the heat equation. Under the above assumption on the interaction strength the decrements fall off at $S \to \infty$ as $1/S^2$. In other words, such a state turns out remarkably stable against noise.

2 Density matrix for an arbitrary spin and its motion equation

We will present the density matrix for spin S as [1, 2]

$$\hat{\rho} = \sum_{l=0}^{2S} \sum_{m=-l}^{l} (-)^m \hat{O}_{lm} P_{l,-m}. \tag{1}$$

Here $\hat{O}_{lm}$ are polarization operators of the rank l and projection m. In the natural spin projection representation their matrix elements are

$$[\hat{O}_{lm}]_{\sigma'\sigma} = \sqrt{\frac{2l+1}{2S+1}}\, C^{S\sigma'}_{S\sigma lm}, \quad \sigma', \sigma = -S, -S+1, \ldots S. \tag{2}$$

where $C^{S\sigma'}_{S\sigma lm}$ is the Clebsch-Gordan coefficient. In particular, at $l=0$ O_{00} is proportional to the $(2S+1)\times(2S+1)$ unit matrix:

$$\hat{O}_{00} = \frac{1}{\sqrt{2S+1}}1. \tag{3}$$

The next polarization operator coincides up to an overall factor with the spin itself:

$$\hat{O}_{1m} = \frac{\sqrt{3}}{\sqrt{S(S+1)(2S+1)}}\hat{S}_m. \tag{4}$$

The polarization moments P_{lm} are in fact the expectation values of the polarization operators in a state described by the density matrix $\hat{\rho}$:

$$P_{lm} = Tr\{\hat{\rho}\hat{O}_{lm}\}. \tag{5}$$

The motion equation for the density matrix is as usual:

$$i\dot{\rho} = [H,\rho]. \tag{6}$$

The Hamiltonian H is conveniently expanded for our problem in the same operators $\hat{O}_{lm}$ used for the parametrization of the density matrix:

$$\hat{H} = -\sum_{lm}(-)^m\hat{O}_{lm}h_{l,-m}; \tag{7}$$

The physical meaning of the parameters $h_{l,-m}$ is clear: h_1 corresponds to cyclic components of a magnetic field, h_2 to those of a gradient of electric field, and so on.

The general commutation relation for the commutator of two arbitrary operators $\hat{O}$ is

$$[\hat{O}_{l_1m_1},\hat{O}_{l_2m_2}] = \sqrt{(2l_1+1)(2l_2+1)}\sum_l(-)^{2S+l}[1-(-)^{l_1+l_2+l}]\left\{\begin{matrix} l_1 & l_2 & l \\ S & S & S\end{matrix}\right\}C^{lm}_{l_1m_1l_2m_2}\hat{O}_{lm}. \tag{8}$$

where $\left\{\begin{matrix} l_1 & l_2 & l \\ S & S & S\end{matrix}\right\}$ is $6j$-symbol. Now, substituting expressions (1), (7) into eq. (6) and using general formula (8) for the commutators, we get the following motion equations for the density matrix parameters:

$$\dot{P}_{lm}(t) = $$
$$= i\sum_{\lambda,l_1,\mu,m_1}[1-(-)^{l+\lambda+l_1}]\sqrt{(2\lambda+1)(2l_1+1)}(-)^{2S+l}\left\{\begin{matrix} l & \lambda & l_1 \\ S & S & S\end{matrix}\right\}C^{lm}_{\lambda\mu l_1m_1}h_{\lambda\mu}(t)P_{l_1m_1}(t). \tag{9}$$

It is convenient to start the investigation of the density matrix relaxation in random fields of arbitrary multipolarities from the simplest case:

3 Relaxation of polarization under magnetic noise

In the special case of a magnetic field Hamiltonian (7) reduces to

$$\hat{H} = -\sum_m(-)^m\omega_{-m}S_m \tag{10}$$

where the usual spin precession frequency ω is introduced instead of h_1:

$$\omega_m = h_{1m}\sqrt{\frac{3}{S(S+1)(2S+1)}} \tag{11}$$

Equation (9) simplifies in this case to

$$\dot{P}_{lm}(t) = -i\sqrt{l(l+1)}\sum_{m_1}\omega_\mu C^{lm}_{1\mu lm_1}P_{lm_1}(t). \tag{12}$$

In other words under magnetic field the parameters of a given multipolarity l change with time independently of other multipolarities. This is only natural. Indeed, the motion equations are generated by the commutation of the Hamiltonian with the density matrix, and spin components entering the magnetic interaction are also the generators of the rotation group under which the tensor operators O_{lm} of a given multipolarity l transform independently. Equation (12) becomes especially simple when the magnetic field does not change its direction, i.e., at $\omega_m = \omega\delta_{m0}$:

$$\dot{P}_{lm} = i\omega m P_{lm}. \tag{13}$$

We wish in particular to find out how the relaxation depends on the spin value S. To this end we have to fix the S-dependence of the parameters h_l entering Hamiltonian (7). Just in the case of a magnetic field there is a natural choice for this dependence: the precession frequency ω of the angular momentum has well-defined classical meaning and is therefore S-independent at least in the classical limit $S\to\infty$. In other words, if we restore explicitly the Planck constant $\hbar$, Hamiltonian (10) should be linear in the angular momentum $\hbar S$.

Now S simply does not enter equation (12) and therefore the evolution of a generalized polarization P_{lm} in arbitrary magnetic fields, including in particular its relaxation in random ones, is independent of the spin S.

It should be kept in mind however, that the S-independence of ω is no more than a natural assumption. Although we will use it, one can well imagine for instance the situation when ω together with g-factor is inversely proportional to S.

The motion equation for the polarization vector in a magnetic field, which constitutes a specific case of equation (12), is well-known. In Cartesian coordinates it is

$$\dot{P}_i = \epsilon_{ijk}P_j\omega_k. \tag{14}$$

Here P_i and ω_k are cartesian coordinates of the polarization and precession frequency, respectively.

We will investigate eq. (14) in the case of isotropic Gaussian noise, in fact assuming that the random magnetic field corresponds to instantaneous kicks with the correlator

$$< \omega_k(t_1)\omega_l(t_2) >= \frac{1}{3}\delta_{kl}\delta(t_1 - t_2)\eta. \tag{15}$$

Here η is related as follows to the typical values of the noise amplitude ω, characteristic time τ, and phase shift at the kicks ϕ:

$$\eta = \omega^2\tau = \phi^2/\tau. \tag{16}$$

One should mention that this problem is only a specific case of a more general one of a rotational Brownian motion of an asymmetric top. That general problem which includes also external random torque and friction has been considered in Ref. [3]. Still the investigation of eq. (14) is appropriate here as a starting point for another general problem we are interested in, that of the relaxation of all momenta of a density matrix in a random field of an arbitrary multipolarity.

The formal solution of (14) can be presented as follows:

$$P_i(t) = \sum_{n=0}^{\infty}(-)^n \int_0^t dt_1 \int_0^{t_1} dt_2 ... \int_0^{t_{n-1}} \epsilon_{ik_1i_1}\omega_{k_1}(t_1)\epsilon_{i_1k_2i_2}\omega_{k_2}(t_2)...\epsilon_{i_{n-1}k_ni_n}\omega_{k_n}(t_n)P_{i_n}(0). \tag{17}$$

Let us average this "solution" over the fluctuations of ω. It can be easily seen that under the assumption (15) a nonvanishing contribution to the sum in rhs of (17) originates from pair correlators of close neighbors only, i.e., of $\omega(t_1)$ with $\omega(t_2)$, $\omega(t_3)$ with $\omega(t_4)$, and so on. Indeed, when pairing for instance $\omega(t_1)$ with $\omega(t_3)$ in (17), one fixes not only $t_3 = t_1$, but $t_2 = t_1$ as well. However, one δ-function in the correlator cannot compensate for two vanishing intervals of integration over time, so this contribution vanishes. After this observation the average value $< P_i(t) >$ can be found from series (17) directly. But we are going to do it by a trick which will be convenient in a more general case.

Let us formally sum series (17) into an integral equation:

$$P_i(t) = P_i(0) - \int_0^t dt_1\epsilon_{ikj}\omega_k(t_1)P_j(0) + \int_0^t dt_1 \int_0^{t_1} dt_2\epsilon_{ikj}\omega_k(t_1)\epsilon_{jlm}\omega_l(t_2)P_m(t_2). \tag{18}$$

When averaging over the fluctuations, the term linear in ω vanishes, and according to the above prescription of pairing the fluctuating fields, the equation becomes

$$< P_i(t) >= P_i(0) + \int_0^t dt_1 \int_0^{t_1} dt_2\epsilon_{ikj}\epsilon_{jlm} < \omega_k(t_1)\omega_l(t_2) >< P_m(t_2) > . \tag{19}$$

This is so-called Bourret's integral equation. Its applicability limits are discussed in Ref. [4], but under our assumption (15) it is evidently true.

We consider expression (15) as a limit of symmetric correlator with a finite correlation interval. So, when substituting (15) into (19), we integrate δ-function according to

$$\int_0^t \delta(t - t')dt' = \frac{1}{2}. \tag{20}$$

Then (19) reduces to

$$< P_i(t) >= P_i(0) - \frac{\eta}{3}\int_0^t dt_1 < P_i(t_1) > \tag{21}$$

with the obvious solution

$$< P_i(t) >= P_i(0)\exp(-\frac{1}{3}\eta t). \tag{22}$$

Certainly, this result, the exponential damping of polarization under gaussian magnetic noise, is well-known.

It is straightforward to generalize the trick applied beyond the pair correlator approximation. Still we assume the Gaussian distribution with the mean square value σ for the phase shifts ϕ at random kicks:

$$W(\phi) = \frac{1}{\sqrt{2\pi\sigma^2}}\exp(-\frac{\phi^2}{2\sigma^2}). \tag{23}$$

Therefore all odd powers of the phase shifts vanish at the averaging, and for their even powers we get

$$< \phi^{2n} >= \sigma^{2n}(2n - 1)!! \tag{24}$$

Clearly, the restriction to the pair correlator corresponds to the assumption $\sigma \ll 1$, or to the case of a "weak" noise.

Now, let $\vec{\nu}$ be the unit vector of the magnetic field (or ω) at a given kick. Then the correlator of the order $2n$ contains the structure

$$\epsilon_{ijk}\nu_j\epsilon_{klm}\nu_l...\epsilon_{rst}\nu_s = (-)^n(\delta_{it} - \nu_i\nu_t) \tag{25}$$

which being averaged over the directions of $\vec{\nu}$ reduces to

$$\frac{2}{3}(-)^n\delta_{it}. \tag{26}$$

In this way we arrive at the following expression for the correlator of the order $2n$:

$$\frac{2}{3\tau}(-)^n\sigma^{2n}(2n - 1)!!\, \delta_{it}\delta(t_1 - t_2)...\delta(t_{2n-1} - t_{2n}) \tag{27}$$

where the characteristic time τ is introduced by dimensional reasons. The same line of reasoning as for the weak noise demonstrates that due to the instantaneous nature of the kicks, we should restrict again to the correlators of the kicks at successive time moments only. In this case as well each δ-function integration brings the factor 1/2. Thus we come to the same integral equation (21), the only difference being in the decrement. Now it is

$$\begin{aligned} \zeta &= -\frac{2}{3\tau}\sum_{n=1}^{\infty}\frac{(-)^n(2n - 1)!!\,\sigma^{2n}}{2^{2n-1}} \\ &= \frac{4}{3\tau}\sqrt{\frac{2}{\pi\sigma^2}}\int_{-\infty}^{\infty}\frac{dx x^2\exp(-2x^2/\sigma^2)}{1 + x^2} \\ &= \frac{4}{3\tau}[1 - \sqrt{\frac{2}{\pi\sigma^2}}\exp(2/\sigma^2)(1 - erf(\sqrt{2/\sigma^2}). \end{aligned} \tag{28}$$

We have introduced here the error function

$$erf(z) = \frac{2}{\sqrt{\pi}} \int_0^z dt \exp(-t^2). \tag{29}$$

In the weak-noise limit $\sigma \ll 1$ this decrement reduces naturally to the previous result $\sigma^2/3\tau$. The strong-noise limit, $\sigma \gg 1$, gives the decrement $\zeta = 4/3\tau$. Naturally, for a strong noise the relaxation time $1/\zeta$ coincides (up to a numerical factor which is in fact a matter of convention) with the characteristic time τ.

The same result (29) for strong noise was obtained earlier by V.V. Sokolov [5].

We are ready now for the discussion of a more general problem:

4 Density matrix relaxation in random fields of arbitrary multipolarities

Rewriting (9) as an integral equation:

$$P(t) = P + i \int_0^t dt_1 \hat{U}(t_1) P(0) - \int_0^t dt_1 \int_0^{t_1} \hat{U}(t_1)\hat{U}(t_2)P(t_2), \tag{30}$$

and averaging it over the weak isotropic gaussian noise we get

$$< P(t) >= P(0) - \int_0^t dt_1 \int_0^{t_1} dt_2 < \hat{U}(t_1)\hat{U}(t_2) >< P(t_2) > . \tag{31}$$

The explicit form of the operator $\hat{U}(t)$ is

$$\hat{U}_{lml_1m_1}(t) = \sum_{\lambda} [1 - (-)^{l+\lambda+l_1}] \sqrt{(2\lambda+1)(2l_1+1)}\, (-)^{2S+l} \left\{ \begin{array}{ccc} l & \lambda & l_1 \\ S & S & S \end{array} \right\} C^{lm}_{\lambda\mu l_1 m_1} h_{\lambda\mu}. \tag{32}$$

The correlator of $h_{\lambda\mu}$ will be written as follows:

$$\begin{aligned} < h_{\lambda\mu}(t) h_{\lambda'\mu'}(t') > &= \frac{1}{2\lambda+1} (-)^{\mu} \delta_{\lambda\lambda'} \delta_{\mu,-\mu'} h^2_{(\lambda)} \tau_\lambda \delta(t-t') = \\ &= \eta_\lambda (-)^{\mu} \delta_{\lambda\lambda'} \delta_{\mu,-\mu'} \delta(t-t'). \end{aligned} \tag{33}$$

Here $h_{(\lambda)}$ is the typical amplitude of fluctuations of multipolarity λ, τ_λ is the corresponding characteristic time. We have introduced also the characteristic decrement η_λ induced by them:

$$\eta_\lambda = \frac{1}{2\lambda+1} h^2_{(\lambda)} \tau_\lambda. \tag{34}$$

Then, using the identity

$$\sum_{\mu, m_1} C^{lm}_{\lambda\mu l_1 m_1} C^{l_1 m_1}_{\lambda,-\mu; l_2 m_2} (-)^{\mu} = (-)^{l+l_1} \sqrt{\frac{2l_1+1}{2l+1}} \delta_{ll_2} \delta_{mm_2} \tag{35}$$

we finally come to an integral equation

$$< P_{lm}(t) >= P_{lm}(0) - \zeta_l \int_0^t dt_1 < P_{lm}(t_1) > \tag{36}$$

which describes the damping of the polarization moments P_{lm} with the decrement

$$\zeta_l = \sum_{\lambda, l'} [1 - (-)^{l+\lambda+l'}] \eta_\lambda (2\lambda+1)(2l'+1) \left\{ \begin{array}{ccc} l & \lambda & l' \\ S & S & S \end{array} \right\}^2 . \tag{37}$$

Let us turn to the simplest case of a purely magnetic noise. At $\lambda = 1$ we get $l' = l$ and

$$\zeta_l = \frac{1}{6} l(l+1)\omega^2 \tau. \tag{38}$$

This result looks extremely natural and can be derived practically without calculations from (13) and (21). Indeed, if according to (13) the instantaneous precession frequency is proportional to the projection m, then it is only natural that the decrement due to the isotropic pair correlator should increase as $l(l+1)$. The overall numerical factor in (38) can be restored from the comparison with (21).

Two features of this result deserve special attention. First, according to (38), higher a polarization moment is, stronger it is damped. Finally, at sufficiently high l (it should be allowed of course by the condition $l \leq 2S$) we will arrive at the strong-noise regime with the decrement $\zeta \sim 1/\tau$. This result is not specific to the magnetic noise, but is valid as well for a noise of any multipolarity, at least for polarization moments with $l \ll 2S$.

Second, if ω is S-independent, the decrement (38) also does not depend on S. In particular nothing happens in the classical limit $S \to \infty$. In this respect as well the situation with higher multipolarities is essentially the same. If the generalized precession frequencies

$$[1 - (-)^{l+\lambda+l_1}] \sqrt{(2\lambda+1)(2l_1+1)}\, (-)^{2S+l} \left\{ \begin{array}{ccc} l & \lambda & l_1 \\ S & S & S \end{array} \right\} C^{lm}_{\lambda\mu l_1 m_1} h_{\lambda\mu}(t) \tag{39}$$

stay finite in the limit $S \to \infty$, as finite are the decrements. At any rate, the S-dependence of decrements is always smooth.

5 Anisotropic Gaussian noise

Since isotropic noise of any multipolarity, as well as arbitrary magnetic one, does not couple polarization moments of different multipolarities, it is instructive to consider anisotropic problem. We will restrict to the simplest nontrivial case of qudrupole noise with one component only:

$$h_{\lambda\mu} = h(t)\delta_{\lambda 2}\delta_{\mu 0}. \tag{40}$$

The explicit form of motion equation (9) for such a field is

$$\begin{aligned} \dot{P}_{lm} &= i6\sqrt{5}[(2S-1)2S(2S+1)(2S+2)(2S+3)]^{-1/2} m h(t) \\ &\cdot \left\{ \sqrt{\frac{[(2S+1)^2 - (l+1)^2][(l+1)^2 - m^2]}{(2l+1)(2l+3)}} P_{l+1,m} + \sqrt{\frac{[(2S+1)^2 - l^2][l^2 - m^2]}{(2l-1)(2l+1)}} P_{l-1,m} \right\} . \end{aligned} \tag{41}$$

In the limit $S \gg l \gg m$ it simplifies to

$$\dot{P}_{lm} = i3\sqrt{5}(2S)^{-3/2} m h(t)(P_{l+1,m} + P_{l-1,m}). \tag{42}$$

We assume again weak Gaussian noise of the typical amplitude h and characteristic time τ, with the δ-function correlator

$$< h(t)h(t') >= \eta\delta(t-t') = h^2\tau\delta(t-t'). \tag{43}$$

Then we arrive at the equation

$$< \dot{P}_{lm} >= -\frac{45\eta m^2}{16S^3}(< P_{l+2,m} > + < 2P_{lm} > + < P_{l-2,m} >). \tag{44}$$

As an initial condition for this equation we choose the pure state with the maximum spin projection S onto the x axis. To obtain the corresponding initial values of the polarization moments P_{lm} we construct first those parameters for the pure state with the same spin projection S onto the z axis and then rotate them by the angle $\beta = \pi/2$ around the y axis. In the state polarized along the z axis the density matrix reduces to $\rho_{\sigma\sigma'} = \delta_{\sigma S}\delta_{\sigma' S}$ and according to (2), (5) we get

$$P_{lm} = \sqrt{\frac{2l+1}{2S+1}} C^{SS}_{SSl0}\delta_{m0}. \tag{45}$$

At $S \gg l$ this expression simplifies to

$$P_{lm} = \sqrt{\frac{2l+1}{2S+1}}\delta_{m0}. \tag{46}$$

Polarization moments, being irreducible tensors, transform under rotation characterized by the Euler angles α, β, γ as follows:

$$P_{lm} = D^l_{mm'}(\alpha,\beta,\gamma)P'_{lm'} \tag{47}$$

where $D^l_{mm'}(\alpha,\beta,\gamma)$ are the Wigner functions. In our case, when $\alpha = \gamma = 0$, $\beta = \pi/2$ and the primed moments corresponding to the polarization along z axis are given by eq. (45), the transformed polarization moments are

$$P_{lm} = \sqrt{\frac{(l-m)!}{(l+m)!}} P^m_l(\cos\frac{\pi}{2})\sqrt{\frac{2l+1}{2S+1}} C^{SS}_{SSl0} \tag{48}$$

where P^m_l are the associated Legendre polinomials. This expression vanishes at odd values of $l-m$. At even $l-m$ it simplifies in the limit $S \gg l \gg 1$ to

$$P_{lm} = (-)^{\frac{l-m}{2}}\sqrt{\frac{2}{\pi S}}. \tag{49}$$

Let us introduce new function $f(l,t) = (-)^{\frac{l-m}{2}} < P_{lm} >$. Equation (44) in terms of f reads

$$\dot{f}(l,t) = \frac{45\eta m^2}{16S^3}[f(l+2,t) - 2f(l,t) + f(l-2,t)] \tag{50}$$

In the limit $l \gg 1$ we come to the heat equation

$$\frac{\partial f(l,t)}{\partial t} = \kappa\frac{\partial^2 f(l,t)}{\partial l^2}, \quad \kappa = \frac{45\eta m^2}{4S^3} \tag{51}$$

with the quite common initial and boundary conditions

$$f(l,t=0) = \sqrt{\frac{2}{\pi S}}, \quad 0 < l < 2S; \tag{52}$$

$$f(0,t) = f(2S,t) = 0. \tag{53}$$

As usual, damping of the function $f(l,t)$ at $t \to \infty$ is controlled by the lowest eigenvalue $\lambda = (\pi/2S)^2$ of the equation

$$\frac{d^2\phi}{dl^2} = -\lambda\phi \tag{54}$$

with the boundary condition $\phi(0) = \phi(2S) = 0$. So, the leading decrement both for the heat equation (54) and for the initial one (51) is

$$\zeta = \kappa(\pi/2S)^2 = \frac{45\pi^2\eta m^2}{16S^4}. \tag{55}$$

Our usual assumption on the S-dependence of noise corresponds to κ staying finite at $S \to \infty$. Then the leading decrement ζ falls off as $1/S^2$. In other words the pure state discussed turns out remarkably stable against noise.

At any rate, the decrement ζ is a smooth function of S as long as the characteristic noise amplitude h (or η) depends on S smoothly.

On the other hand, in the strong-noise limit $\zeta\tau \gg 1$ it can be shown by simple analysis that the leading decrement tends roughly to $1/\tau$ as it was in the isotropic case.

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