The Limit Diffusion Mechanism of Relaxation for Spin Systems

by

Lech Papież*

Dublin Institute for Advanced Studies
10 Burlington Road
Dublin 4, Ireland

Abstract

The diffusion limit theorem for stochastic differential equations is applied to analyse the dynamical evolutions of spin systems. Bloch equations are derived and the stability of asymptotic evolutions is proved. The theory is applied to nuclear magnetic relaxation of two spins.

* On leave of absence from Silesian University, Katowice, Poland.

Introduction and Motivation

Any effective method of statistical physics uses somewhere certain assumptions which are essentially of a stochastic nature. In the theory of nonequilibrium statistical mechanics advocated by Kubo [1-3] and Van Kampen [4,5], where the time evolution of physical systems is postulated on the basis of stochastic dynamical equations (stochastic Liouville equations), these assumptions are expressed more openly. However, derivations of kinetic equations from stochastic Liouville equations [1-10] have, in the past, caused much confusion. It was not clear that assumptions to neglect some correlations, necessary for these derivations, were justified [6-10] (this subtle problem was exhaustively discussed, for example, in [5]). By analogy with the deterministic case one may expect, however, that the Van Hove limit [11,12] is the proper method for the analysis of stochastic Liouville equations, and clearly should lead to kinetic equations. Fortunately, a mathematical theory exists (the so-called asymptotic theory of stochastic multiplicative equations, which started in 1966 with the Stratonovich [13] and Khasminskii [14] paper) which is appropriate for such analysis. We are thus prompted to check how such a theory will work for some simple physical systems. Spin systems, with their finite degrees of freedom and well-established relaxation results are specially good candidates. Thus the object of this paper is to show that the asymptotic theory of stochastic, multiplicative equations [13-17] applies to quantum evolution for spin systems in a random environment, and that it allows us to derive rigorously kinetic equations (Bloch equations) and to determine transport coefficients (relaxation times). Moreover, because our considerations are based on mathematical results, we are able to dig out those assumptions which are of an "essentially stochastic nature", under which the relaxation in spin systems appears.

The paper is constructed in the following way:
Firstly, we state the general problem of the spin system evolution in a random environment. Then, in Section 2, we describe a particular example of such a system for which we aim to derive explicit results: and, in Section 3, we transform its evolution equation into multiplicative, stochastic form. In Section 4 we quote the results which are later,

in the more theoretical Sections 5 and 6, rigorously justified. Section 5 is thus devoted to the statement of the relevant diffusion limit theorem which allows us to obtain the time evolution of spin systems, while in Section 6 the general consequences of this evolution are investigated. Eventually, in Section 7, we make the final calculations for the system of two spins.

1. Statement of the problem

We aim to derive the Bloch equations and relaxation times for nuclear magnetism from stochastic dynamical equations. Thus the definition of the system under consideration is the same as in the Redfield [6] and Kubo [1,2] papers. The Hamiltonian of the system of spins and their molecular environment (bath) can be written in the form

$$H = H(S) + H(S,q) \tag{1}$$

where H(S) depends only on the spin variables S, and H(S,q) is the energy of interaction of the spin S and the molecular q degrees of freedom. The stochasticity of H(S,q) is the result of the random variation with time t ($t \geqslant 0$) of coordinates q (if such probabilistic assumptions are made in consideration of the quantum evolution the theory is called "semiclassical" [6,18,19,20]). From quantum mechanical laws we have the evolution of our spin-bath system given by

$$\frac{d\rho(t)}{dt} = \frac{1}{i}\left[H, \rho(t)\right], \qquad \rho(0) = \rho_0, \quad t \geqslant 0 \tag{2}$$

where ρ is the density matrix and H is the Hamiltonian (1). The equation (2) is a stochastic one and its solution $\rho(t)$ is a stochastic process which determines the evolution of the spin-bath system, while the average $E\,\rho(t)$ (over bath) may be interpreted as the density matrix of the spin subsystem.

Our investigations of the quantum evolution (2) will proceed in the sense of the weak coupling limit [11,12,21]. This treatment of the dynamical evolution equations, which leads to derivations of kinetic equations, started with Van Hove's papers [11,12], and is now one of the most powerful methods of nonequilibrium statistical mechanics [21].

In our case it means that instead of equation (2) we should postulate

$$\frac{d\rho^{\varepsilon}(t)}{dt} = \frac{\varepsilon}{i}\left[H, \rho^{\varepsilon}(t)\right], \qquad \rho^{\varepsilon}(0) = \rho_0, \quad t \geqslant 0 \tag{3}$$

where H is the same Hamiltonian as before, and treat as the (macroscopic) physical evolution that matrix $\rho^{\varepsilon}(t)$ which is the solution of (3) for $\varepsilon \to 0$ and $0 \leqslant \varepsilon^2 t = \tau \leqslant \tau_0$, where τ_0 is arbitrary. ε should be interpreted here as a scaling parameter, rather than any established physical quantity. To realize this let us express, the time-dependent Hamiltonian H(t) given in (1) as

$$H(t) = H^{\varepsilon}(t/\varepsilon), \tag{4}$$

instead of postulating ad hoc the presence of ε in (3). Then (2) yields

$$\frac{d\rho^{\varepsilon}(t)}{dt} = \frac{\varepsilon}{i}\left[H^{\varepsilon}(t), \rho^{\varepsilon}(t)\right], \qquad \rho^{\varepsilon}(0) = \rho_0, \quad t \geqslant 0 \tag{5}$$

where $\rho^{\varepsilon}(t/\varepsilon) = \rho(t)$. If we now demand the same stochastic characteristics for $H^{\varepsilon}(t)$ as we have required earlier for H(t), then the solution $\rho^{\varepsilon}(t)$ of (5) for $\varepsilon \to 0$ and $0 \leqslant \varepsilon^2 t = \tau \leqslant \tau_0$ will be the same as the solution obtained using the weak coupling postulate. In this sense the weak coupling postulate is equivalent to time scaling.

2. The system of two spins in a random environment

Here, we restrict our considerations to a simple system of two spins contained in a molecule placed in the random environment and interacting only through dipole-dipole interactions. We assume also the absence of the external magnetic field, i.e. H(S) = 0 (let us notice that H(S,q) is the part of Hamiltonian (1) responsible for the relaxation). Using the example of this system, we are able to show how the method works while the explicit expressions are as simple as possible. The Hamiltonian H(t) = H(S,q) of the spin dipole-dipole interaction is in this case [6,18-20]

$$H(t) = \sum_{k=-2}^{2} F^{k}(t)\, V^{k}, \tag{6}$$

and the $F^k(t)$ are equal to

$$F^k(t) = \left(\frac{6\pi}{80}\right)^{1/2} \gamma^2 \hbar^2 r^{-3} (-1)^k Y_2^{-k}\left(\Theta(t), \varphi(t)\right), \quad k = 0, \pm 1, \pm 2, \qquad (7)$$

where the Y_2^{-k} are second order spherical harmonics dependent on Θ and φ which denote angles determining the orientation of a vector $\bar{r}$ "joining" two spins in a molecule, and r is the length of $\bar{r}$. The second order tensor operators V^k appearing in (6) are [19,20]

$$V^0 = -\left(\frac{8}{3}\right)^{1/2}\left[S_3 \otimes S_3 - \frac{1}{4}\left(S_1 \otimes S_{-1} + S_{-1} \otimes S_1\right)\right] =$$

$$= -\frac{2}{\sqrt{6}}\left(2 S_3 \otimes S_3 - S_1 \otimes S_1 - S_2 \otimes S_2\right), \qquad (8)$$

$$V^{\pm 1} = \pm\left(S_3 \otimes S_{\pm 1} + S_{\pm 1} \otimes S_3\right) = \pm\left(S_3 \otimes S_1 + S_1 \otimes S_3\right) + i\left(S_3 \otimes S_2 + S_2 \otimes S_3\right),$$

$$V^{\mp 2} = -S_{\pm 1} \otimes S_{\pm 1} = -\left(S_1 \otimes S_1 - S_2 \otimes S_2\right) \mp \left(S_2 \otimes S_1 + S_1 \otimes S_2\right)$$

where S_1, S_2 and S_3 denote the Pauli spin matrices

$$S_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \qquad S_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \qquad S_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \qquad (9)$$

where $S_{\pm 1} = S_1 \pm i S_2$.

According to the statement of the problem, the $F^k(t)$ are random variables for any $t \geqslant 0$. Thus, to define our system we must determine the probabilistic properties of stochastic processes $F^k(t)$. Let us mention the most obvious of them. From (7) and properties of spherical harmonics Y_2^k it follows that

(1F) $F^k(t)$ are bounded random variables for any $t \geqslant 0$.

The natural requirement that the molecular environment is homogeneous

in time, and isotropic, implies:

(2F) $F^k(t)$ are stationary processes in a wide sense, i.e. $E F^k(t) = C$ (without making our considerations less general we may choose $C = 0$), and $R_{k,k'}(t_1,t_2) = E F^k(t_1) F^{k'*}(t_2) = R_{k,k'}(t_1 - t_2)$,

and (3F) the correlation matrix $R_{k,k'}(s)$ for $F^k(t)$ processes is diagonal, i.e.

$$R_{k,k'}(s) = \delta_{k,k'} R_k(s) \qquad \text{where} \quad R_k(s) = E F^k(s) F^{k*}(0).$$

(2F) was always assumed in physical models of nuclear magnetic relaxation [6,18-20,22]. (3F) follows from properties of spherical harmonics Y_2^k and the isotropic initial distribution of the orientation of a molecule carrying two spins (because of homogenity in time this isotropic distribution is preserved during the motion). This was shown, for example, by Ford, Lewis and McConnell ([23], Appendix) and by Hubbard [24]. Further, the investigations of all specific, stochastic models of rotational motions of molecules demonstrate that [25]

(4F) correlation functions $R_k(s) = E F^k(s) F^{k*}(0)$ are real, i.e.

$$R_k(s) = R_k^*(s) \quad \text{, which implies} \quad R_k(s) = R_k^*(s) = R_k(-s) = R_{-k}(s).$$

At last we know that any nondegenerate stochastic process, in contrast with a deterministic one, must forget, to some extent, its previous evolution. Thus

(5F) Processes $F^k(t)$ acquire, quickly enough, independence of the timely evolution from the previous one.

The precise formulation of the condition (5F) we postpone to our later discussion of the mathematical theory of stochastic, multiplicative differential equations.

3. Reduction of the density matrix equation to a linear differential equation in Euclidean space

Let us construct the auxiliary, real, Euclidean space R_s in which the quantum evolution equation (3) assumes, in the case of the system of two spins, the form of a linear, differential equation. To this end let us define the scalar product (,) for the basis set of vectors $\{S_i \otimes S_j\}_{i,j=1}^{4}$ by

$$\left(S_i \otimes S_j, S_k \otimes S_l\right) = \frac{1}{4} Tr\left(\left(S_i \otimes S_j\right) \cdot \left(S_k \otimes S_l\right)\right), \qquad (10)$$

$$i, j, k, l = 1, 2, 3, 4,$$

and the $F^k(t)$ are equal to

$$F^k(t) = \left(\frac{6\pi}{30}\right)^{1/2} \gamma^2 \hbar^2 r^{-3} (-1)^k \, Y_2^{-k}\left(\theta(t), \varphi(t)\right), \quad k = 0, \pm 1, \pm 2, \qquad (7)$$

where the Y_2^{-k} are second order spherical harmonics dependent on θ and φ which denote angles determining the orientation of a vector $\bar{r}$ "joining" two spins in a molecule, and r is the length of $\bar{r}$. The second order tensor operators V^k appearing in (6) are [19,20]

$$V^0 = -\left(\frac{8}{3}\right)^{1/2} \cdot \left[S_3 \otimes S_3 - \frac{1}{4}\left(S_1 \otimes S_{-1} + S_{-1} \otimes S_1\right)\right] =$$

$$= -\frac{2}{\sqrt{6}} \left(2 S_3 \otimes S_3 - S_1 \otimes S_1 - S_2 \otimes S_2\right), \qquad (8)$$

$$V^{\pm 1} = \pm \left(S_3 \otimes S_{\pm 1} + S_{\pm 1} \otimes S_3\right) = \pm \left(S_3 \otimes S_1 + S_1 \otimes S_3\right) + i \left(S_3 \otimes S_2 + S_2 \otimes S_3\right),$$

$$V^{\mp 2} = - S_{\pm 1} \otimes S_{\pm 1} = -\left(S_1 \otimes S_1 - S_2 \otimes S_2\right) \mp \left(S_2 \otimes S_1 + S_1 \otimes S_2\right)$$

where S_1, S_2 and S_3 denote the Pauli spin matrices

$$S_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \qquad S_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \qquad S_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \qquad (9)$$

where $S_{\pm 1} = S_1 \pm i S_2$.

According to the statement of the problem, the $F^k(t)$ are random variables for any $t \geqslant 0$. Thus, to define our system we must determine the probabilistic properties of stochastic processes $F^k(t)$. Let us mention the most obvious of them. From (7) and properties of spherical harmonics Y_2^k it follows that

(1F) $F^k(t)$ are bounded random variables for any $t \geqslant 0$.

The natural requirement that the molecular environment is homogeneous

in time, and isotropic, implies:

(2F) $F^k(t)$ are stationary processes in a wide sense, i.e. $E F^k(t) = C$ (without making our considerations less general we may choose $C = 0$), and $R_{k,k'}(t_1, t_2) = E F^k(t_1) F^{k'*}(t_2) = R_{k,k'}(t_1 - t_2)$,

and (3F) the correlation matrix $R_{k,k'}(s)$ for $F^k(t)$ processes is diagonal, i.e.

$$R_{k,k'}(s) = \delta_{k,k'} R_k(s) \qquad \text{where} \qquad R_k(s) = E F^k(s) F^{k*}(0).$$

(2F) was always assumed in physical models of nuclear magnetic relaxation [6,18-20,22]. (3F) follows from properties of spherical harmonics Y_2^k and the isotropic initial distribution of the orientation of a molecule carrying two spins (because of homogenity in time this isotropic distribution is preserved during the motion). This was shown, for example, by Ford, Lewis and McConnell ([23], Appendix) and by Hubbard [24]. Further, the investigations of all specific, stochastic models of rotational motions of molecules demonstrate that [25]

(4F) correlation functions $R_k(s) = E F^k(s) F^{k*}(0)$ are real, i.e.

$$R_k(s) = R_k^*(s) \quad \text{, which implies} \quad R_k(s) = R_k^*(s) = R_k(-s) = R_{-k}(s).$$

At last we know that any nondegenerate stochastic process, in contrast with a deterministic one, must forget, to some extent, its previous evolution. Thus

(5F) Processes $F^k(t)$ acquire, quickly enough, independence of the timely evolution from the previous one.

The precise formulation of the condition (5F) we postpone to our later discussion of the mathematical theory of stochastic, multiplicative differential equations.

3. Reduction of the density matrix equation to a linear differential equation in Euclidean space

Let us construct the auxiliary, real, Euclidean space R_s in which the quantum evolution equation (3) assumes, in the case of the system of two spins, the form of a linear, differential equation. To this end let us define the scalar product (,) for the basis set of vectors $\{S_i \otimes S_j\}_{i,j=1}^{4}$ by

$$\left(S_i \otimes S_j, S_k \otimes S_l\right) = \frac{1}{4} Tr\left(\left(S_i \otimes S_j\right) \cdot \left(S_k \otimes S_l\right)\right), \qquad (10)$$

$$i,j,k,l = 1,2,3,4,$$

where S_1, S_2 and S_3 are spin Pauli matrices (9), $S_4 = I$ is the identity 2 x 2 matrix, and $\cdot$ denotes the matrix multiplication. From the above definition it follows that

$$\left(S_i \otimes S_j , S_k \otimes S_l \right) = \delta_{ik} \delta_{jl} , \qquad i,j,k,l = 1,2,3,4, \tag{11}$$

and in the standard notation for basis vectors, i.e. when

$$S_1 \otimes S_1 = e_1, \quad S_1 \otimes S_2 = e_2, \ \dots, \ S_4 \otimes S_4 = e_{16}, \tag{12}$$

we may write instead of (11)

$$\left(e_i , e_j \right) = \delta_{ij} , \qquad i,j = 1, \dots, 16. \tag{13}$$

The set $\{ e_i \}_{i=1}^{16}$ may serve therefore as the ortonormal basis of the real, sixteen — dimensional Euclidean space R_s^{16}, and any vector $z \in R_s^{16}$, $z = \sum_{i=1}^{16} z_i e_i$, where z_i are real, may be thought of as a 4 x 4 hermitian matrix which is a linear combination of basis matrices $\{ S_i \otimes S_j \}_{i,j=1}^{4}$. Since vectors from R_s^{16} are identical with 4 x 4 hermitian matrices, the scalar product (z,y) of z and y may be written as

$$(z,y) = \frac{1}{4} \mathrm{Tr} \left(z \cdot y \right); \tag{14}$$

hence immediately

$$z_i = \left(z, e_i \right) = \frac{1}{4} \mathrm{Tr} \left(z \cdot e_i \right) \tag{15}$$

(in particular $z_{16} = \frac{1}{4} \mathrm{Tr}\,(z)$), (15')

and

$$|z|^2 = \frac{1}{4} \mathrm{Tr} \left(z^2 \right). \tag{16}$$

Thus we recognize that any density matrix ρ for the system of two spins is a vector $z \in R_s^{16}$ for which the last component z_{16} is equal to $\frac{1}{4}$ and $|z| \leq \frac{1}{2}$. The other components z_i, i = 1, ..., 15 of a vector z have by (15) the interpretation of quantum averages of observables e_i defined

in (12). Moreover, the Hamiltonian $H(t)$ of our problem, defined by (6), (7), (8), may be considered as a stochastic process with values in R_s^{16}. Therefore, since the commutator $[\ ,\ ]$ in (3) is the relevant algebraic operation in R_s^{16}, the evolution equation (3), with Hamiltonian (6), is the linear (multiplicative) stochastic differential equation in R_s^{16}. More specifically we may write

$$\frac{d}{dt} \left(\sum_{i=1}^{16} z_i^{\varepsilon} e_i \right) = \frac{\varepsilon}{i} \left[H(t), \sum_{j=1}^{16} z_j^{\varepsilon} e_j \right] = \frac{\varepsilon}{i} \left[\sum_{k=-2}^{2} F^k(t) V^k, \sum_{j=1}^{16} z_j^{\varepsilon} e_j \right] =$$
$$= \varepsilon \sum_{j=1}^{16} \sum_{k=-2}^{2} F^k(t) \left(\frac{1}{i} \left[V^k, e_j \right] \right) z_j^{\varepsilon}, \qquad \sum_{i=1}^{16} z_i^{\varepsilon}(0) e_i = z \in R_s^{16}. \tag{17}$$

Taking the scalar product of the left and the right hand side of (17) with basic vector e_l, we obtain

$$\frac{d}{dt} z_l^{\varepsilon} = \varepsilon \sum_{j=1}^{16} \sum_{k=-2}^{2} F^k(t) A_{lj}^k z_j^{\varepsilon} , \qquad z_l^{\varepsilon}(0) = z_l , \qquad l = 1, \dots, 16 \tag{18}$$

where A_{lj}^k are defined by

$$A_{lj}^k = \frac{1}{i} \left(e_l, \left[V^k, e_j \right] \right), \qquad l,j = 1, \dots, 16; \quad k = 0, \pm 1, \pm 2. \tag{19}$$

The hermiticity of V^k, e_i and e_j and the definition of the scalar product in R_s^{16} imply that matrices $\{ A^k \}_{k=-2}^{2}$ are skew-symmetric, i.e.

$$\left(A^k \right)^T = - A^k. \tag{20}$$

In a vector notation (18) takes the form

$$\frac{d z^{\varepsilon}(t)}{dt} = \varepsilon \sum_{k=-2}^{2} F^k(t) A^k z^{\varepsilon}(t), \qquad z^{\varepsilon}(0) = z \in R_s^{16}. \tag{21}$$

Finally we find by substitution of (8) and (12) into (19) that

$$\left(A^k \right)^* = (-1)^k A^{-k}. \tag{22}$$

The combination of (22) with the analogous relation for spherical harmonics, i.e.

$$Y_2^{k*}(t) = (-1)^k \, Y_2^{-k}(t) \quad \text{or} \quad F^{k*}(t) = (-1)^k \, F^{-k}(t) \tag{23}$$

ensures that the operator $B(t) = \sum_{k=-2}^{2} F^k(t) A^k$ is a real, linear operator from R_S^{16} into R_S^{16}.

This concludes the transformation of the time evolution of our physical system into the problem of linear, multiplicative differential equation in the real, Euclidean space R_S^{16}.

4. Results

To complete the discussion of our example, we now quote results which we will justify rigorously later. All these results follow from the solution $\rho^\varepsilon(t)$ which we are able to obtain (for $\varepsilon \to 0$ and $0 \leq \varepsilon^2 t = \tau \leq \tau_0$, $\rho^\varepsilon(t) \to \rho(\tau)$) for the problem (3) when H is the Hamiltonian of the system given by (6), (7) and (8). This solution determines completely the evolution of the spin-bath system on the macroscopic time-scale. Let $\rho_S(\tau)$ denote the evolution of the density matrix of the spin subsystem, i.e. $\rho_S(\tau) = E\rho(\tau)$, and let $\langle G \rangle$ denote the global average (over the spin subsystem and its random environment - bath) of the spin observable G, i.e. $\langle G \rangle = Tr\, E(\rho \cdot G) = Tr(\rho_S \cdot G)$. The results are:

(i) Conditions $Tr\,\rho_S(\tau) = 1$ and $Tr\,\rho_S^2(\tau) \leq 1$ are fulfilled for any $\tau \in [0, \tau_0]$ if they are true for $\tau = 0$;

(ii) The observable $x_0(S_1 \otimes S_1 + S_2 \otimes S_2 + S_3 \otimes S_3)$, x_0-real parameter, is invariant during the evolution of the system. Parameter x_0 is determined only by the initial conditions of the spin subsystem;

(iii) The density matrix of spin subsystem $\rho_S(\tau)$ for $\tau \to \infty$ relaxes to the limit (equilibrium) density matrix ρ_{eq} given by

$$\rho_{eq} = \begin{pmatrix} \frac{1}{4}+x_0 & 0 & 0 & 0 \\ 0 & \frac{1}{4}-x_0 & 2x_0 & 0 \\ 0 & 2x_0 & \frac{1}{4}-x_0 & 0 \\ 0 & 0 & 0 & \frac{1}{4}+x_0 \end{pmatrix} \tag{24}$$

where x_0 is as in (ii). We notice that the dependence on temperature of the bath is absent here as the part H(S) of the Hamiltonian is assumed to be 0;

(iv) The evolutions of $\langle S^z \rangle$ and $\langle S^{\pm 1} \rangle$, where S^z and $S^{\pm 1}$ denote, respectively, z and ± 1 components of the total spin operator of two spins

$$S^z = \frac{1}{2}\left(S_3 \otimes S_4 + S_4 \otimes S_3\right) \tag{25}$$

and

$$S^{\pm 1} = \frac{1}{2}\left[\left(S_1 \otimes S_4 + S_4 \otimes S_1\right) \pm i\left(S_2 \otimes S_4 + S_4 \otimes S_2\right)\right], \tag{26}$$

are given by

$$\langle S^z \rangle(\tau) = \langle S^z \rangle(0) \cdot \exp\left(-\frac{\tau}{T_1}\right) \tag{27}$$

and

$$\langle S^{\pm 1} \rangle(\tau) = \langle S^{\pm 1} \rangle(0) \cdot \exp\left(-\frac{\tau}{T_2}\right). \tag{28}$$

T_1 and T_2 denote, respectively, longitudinal and perpendicular relaxation times, which will be derived in Section 7. Because we assumed H(S) = 0 we should compare T_1 and T_2 with the relaxation times for the case of extreme narrowing which were obtained earlier [6,18-20,22]. It is easy to verify that our results are identical with those known earlier [20,22].

5. The solutions of stochastic dynamical equations for spin systems

Let us consider the stochastic multiplicative equation in R^n of the kind

$$\frac{dz^\varepsilon(t)}{dt} = \varepsilon \sum_{k=-l}^{l} F^k(t) A^k z^\varepsilon(t), \quad z^\varepsilon(0) = z \in R^n, \quad t \geq 0 \tag{29}$$

where $F^k(t)$, $t \geq 0$, $k = 0, \pm 1, \ldots, \pm l$ are components of a given stochastic process F(t) on a probability space $(\Omega, \mathcal{F}, P)$ (we shall denote integration over Ω relative to P, i.e. the bath average, by E), and $\{A^k\}_{k=-l}^{l}$ are given constant n x n matrices. This equation is analogous to (21) except that n and l are now arbitrary natural numbers (in (21)

$n = 16$ and $\ell = 2$); it describes spin systems which are more complex than than those considered in our example. They must, however, be such as to satisfy conditions (20), (22), (23) and (1F) - (5F) for any $k = 0, \pm 1, \ldots, \pm \ell$.

In order to obtain the solution for (29) in a rigorous manner we must make the condition (5F) more exact. We assume that processes $F^k(t)$ acquire the independence of the present evolution from the previous one in the sense, that:

(5F') correlation functions $R_k(s)$ converge to zero for $s \to \infty$ fast enough to ensure that

$$\int_0^\infty | s R_k(s) | \, ds < C, \tag{30}$$

and (5F") process F(t) satisfies the mixing condition [14-17,26-28] i.e.

$$\sup_{t \geq 0} \sup_{\substack{A \in \mathcal{F}_{t+s}^\infty \\ B \in \mathcal{F}_0^t}} | P(A|B) - P(A) | = \alpha(s) \xrightarrow{s \to \infty} 0 \tag{31}$$

where $\mathcal{F}_s^t \subset \mathcal{F}$, $0 \leq s \leq t \leq \infty$ denotes an σ-algebra of events generated on Ω by $F(u)$, $s \leq u \leq t$ and $\alpha(s)$ fulfils

$$\int_0^\infty \alpha^{1/2}(s) \, ds < \infty . \tag{32}$$

As we said earlier, the property of "losing the memory" by process F(t) is obvious. However, the questions in what sense, and how fast, it is lost are open ones. In this context (5F') and (5F") should be seen mainly as sufficient conditions for $F^k(t)$ under which the relaxation in the considered systems appears.

Remark 1

It is known [26,27] that for Gaussian stationary processes the mixing condition is equivalent to the complete regularity condition [17,26,27], which allows us to estimate the correlation function. Therefore, by restricting ourselves to Gaussian processes, we may get rid of the condition (5F').

Remark 2

Ergodic Markov processes $F^k(t)$ are examples of processes for which (5F") is satisfied [16].

Let us state now some simple consequences of (20), (1F) - (5F'), (5F").

Lemma 1

The quantities J_k defined as

$$J_k = \int_0^\infty R_k(s) \, ds \tag{33}$$

are equal to

$$\lim_{T \to \infty} \frac{1}{T} \int_0^T \int_0^s R_k(s-s') \, ds' ds , \tag{34}$$

and (34) converges to J_k for $T \to \infty$ as $\frac{1}{T}$.

Proof

Lemma 1 is implied by (5F') because, thanks to the partial integration of (34), we have

$$\lim_{T \to \infty} \frac{1}{T} \int_0^T \int_0^s R_k(s-s') \, ds' ds = \lim_{T \to \infty} \int_0^T (T-s) R_k(s) \, ds =$$
$$= J_k - \lim_{T \to \infty} \frac{1}{T} \int_0^T s R_k(s) \, ds . \tag{35}$$

We notice that by (4F) J_k are positive, and $J_k = J_{-k}$, $k = 0, \pm 1, \ldots, \pm \ell$.

Corrolary

Applying (3F), (20), and Lemma 1 we find that following limits

$$b_i(z) = \lim_{T \to \infty} \frac{1}{T} \int_0^T \int_0^s \sum_{j=1}^n E\Big(\sum_{k=-\ell}^{\ell} F^k(s) A^k z\Big)_j \frac{\partial}{\partial z_j} \Big(\sum_{k'=-\ell}^{\ell} F^{k'}(s') A^{k'} z\Big)_i^* \, ds' ds \tag{36}$$

and

$$c_{ij}(z) = \lim_{T \to \infty} \frac{1}{T} \int_0^T \int_0^s E\Big(\sum_{k=-\ell}^{\ell} F^k(s) A^k z\Big)_i \Big(\sum_{k'=-\ell}^{\ell} F^{k'}(s') A^{k'} z\Big)_j^* \, ds' ds \tag{37}$$

exist uniformly in z. In vector notation, (36) and (37) may be put into the form

$$b(z) = [b_i(z)] = \sum_{k=-L}^{L} J_k (A^k)^\dagger A^k z = -Dz \tag{38}$$

and
$$C(z) = [c_{ij}(z)] = \sum_{k=-L}^{L} J_k A^k z z^T (A^k)^\dagger \tag{39}$$

where $(A^k)^\dagger$ denotes the hermitian conjugate of A^k.

Lemma 2

Matrices $D = \sum_{k=-L}^{L} J_k (A^k)^\dagger A^k$ and $C(z)$ are nonnegative definite.

Proof

Matrices D and $C(z)$ are real, and for any $\xi \in R^n$ we have

$$\xi^T D \xi = \sum_{k=-L}^{L} J_k (A^k \xi)^\dagger A^k \xi \geq 0 \tag{40}$$

and
$$\xi^T C(z) \xi = \sum_{k=-L}^{L} J_k (\xi^T A^k z)(\xi^T A^k z)^\dagger \geq 0, \tag{41}$$

as the J_k are positive.

Let us notice here that matrices D and $C(z)$, though nonnegative definite, may be degenerate since the A^k are skew-symmetric.

Now, let us recall the limit theorems for stochastic multiplicative differential equations [13-17]. For our case the relevant versions of these theorems are given in the papers of Papanicolaou and Varadhan [15] and Papanicolaou and Kohler [16]. It is not difficult to recognize that our problem is analogous to the one considered in Theorem 3 of [15], and that after similar transformation, the general, abstract Theorem 1 of [15] will apply. It would be more convenient, however, to recall the theorem of Papanicolaou and Kohler [16] (PK Theorem). Because $F^k(t)$ are bounded and since the equation (29) is linear (and not explicitely dependent on the macroscopic time scale) all the boundedness conditions needed for PK Theorem are satisfied. Moreover, the existence of the drift $b_i(z)$ and the diffusion $c_{ij}(z)$ coefficients, in the sense of limits (35), (36) and (37), and their explicit forms (38) and (39), ensure the fulfillment of the further conditions of the PK Theorem. Thus the PK Theorem applies. Its relevance for the formulation of our problem may be expressed as follows:

Theorem

Let $z^\varepsilon(t) = \tilde{z}^\varepsilon(\tau)$, $0 \leq \varepsilon^2 t = \tau \leq \tau_0$, $\varepsilon \in (0,1]$ be the process defined by equation (29). Let (1F) - (5F'), (5F"), (20), (22) and (23) hold. Then the process $\tilde{z}^\varepsilon(\tau)$ converges weakly as $\varepsilon \to 0$ ($t \to \infty$ and $0 \leq \varepsilon^2 t = \tau \leq \tau_0$, τ_0 arbitrary) to the diffusion Markov process $z(\tau)$ with infinitesimal generator L defined by

$$L = \sum_{i,j=1}^{n} c_{ij}(z) \frac{\partial}{\partial z_i \partial z_j} + \sum_{i=1}^{n} b_i(z) \frac{\partial}{\partial z_i} \tag{42}$$

where $b_i(z)$ and $c_{ij}(z)$ are, respectively, drift and diffusion coefficients given by (38) and (39).

Remark 3

Theorems also exist ([17], theorems 2 and 3, chapter 7, § 3) which allow us to determine the behaviour of the solution $z^\varepsilon(t)$ of the above problem for $t \in [0, \tau_0/\varepsilon]$. It turns out that for such time-intervals process $z^\varepsilon(t)$ does not move significantly from its initial position [17,28]. Changes in $z^\varepsilon(t)$ for $t \in [0, \tau_0/\varepsilon]$ are of the order $\varepsilon^{1/2}$, and not of the order 1 as expected for $t \in [0, \tau_0/\varepsilon^2]$ from the above theorem, [17,28]. This explains, to some extent, why we have formulated our problem in the Van Hove limit.

6. Consequences of the diffusion limit solution for spin systems

For further discussion it would be more convenient to represent the diffusion Markov process $z(\tau)$ with drift vector $b(z)$ and diffusion matrix $C(z)$, as the solution of the Ito type stochastic equation [29,30]. We have

$$dz(\tau) = b(z(\tau))\, d\tau + \sigma(z(\tau))\, dW_\tau, \quad z(0) = z \in R^n, \quad \tau \geq 0 \tag{43}$$

where $\sigma(z)$ is a square root matrix of $C(z)$ (because $C(z)$ is a nonnegative definite matrix it has a unique square root $\sigma(z)$ [30]), and W_τ is a standard, n-dimensional Wiener process.

Lemma 3

The mean value $E\,z(\tau)$ of the process $z(\tau),\ \tau \geq 0$ is contained in $K_{r,0} = \{z \in R^n : |z| \leq r\}$ where $r = |z(0)|$, and $z(0)$ denotes the starting point of the evolution.

Proof

Let us consider the function

$$v(z) = |z|^{2p} = \left(\sum_{i=1}^{n} z_i^2\right)^p . \tag{44}$$

Because of the following identities

$$\frac{\partial}{\partial z_i} |z|^{2p} = 2p z_i |z|^{2(p-1)} \tag{45}$$

and
$$\frac{\partial}{\partial z_i \partial z_j} |z|^{2p} = 2p \delta_{ij} |z|^{2(p-1)} + 4p(p-1) z_i z_j |z|^{2(p-2)} \tag{46}$$

we have

$$\begin{aligned} L v(z) = {} & 2p \sum_{j=1}^{n} \left(\sum_{k=-l}^{l} J_k \sum_{m,m'=1}^{n} A^k_{jm} A^{k*}_{jm'} z_m z_{m'} \right) |z|^{2(p-1)} \\ & + 4p(p-1) \left(\sum_{k=-l}^{l} J_k \sum_{i,j,m,m'=1}^{n} A^k_{im} A^{k*}_{jm'} z_m z_{m'} z_i z_j \right) |z|^{2(p-2)} \\ & + 2p \sum_{j=1}^{n} \left(\sum_{k=-l}^{l} J_k \sum_{m',m=1}^{n} A^k_{m'j} A^{k*}_{jm} z_m z_{m'} \right) |z|^{2(p-1)} . \end{aligned} \tag{47}$$

The first and the third terms in (47) cancel, since matrices A^k are skew-symmetric. Therefore (47) yields

$$\begin{aligned} L v(z) & = 4p(p-1) \sum_{k=-l}^{l} J_k \, z^T A^k z \, z^T (A^k)^{\dagger} z \, |z|^{2(p-2)} \\ & = 4p(p-1) \, z^T C(z) z \, |z|^{2(p-2)} , \end{aligned} \tag{48}$$

and by nonnegative definitness of C(z) we obtain

$$L v(z) \leq 0 \qquad \text{for} \qquad 0 \leq p \leq 1 . \tag{49}$$

By Ito's formula we have also

$$d v(z(\tau)) = L v(z(\tau))\, d\tau + \frac{\partial v(z(\tau))}{\partial z} \sigma(z(\tau))\, dW_\tau \tag{50}$$

Applying the average to the last expression we obtain

$$E\, d v(z(\tau)) = E\, L v(z(\tau)) \leq 0 \qquad \text{for} \qquad 0 \leq p \leq 1 . \tag{51}$$

Hence, for $p = \frac{1}{2}$, $v(z) = |z|$.

$$|E\, z(\tau)| \leq E\,|z(\tau)| \leq E\,|z(0)| = |z(0)| \tag{52}$$

Thus Lemma 3 is true, and evidently also it implies that the inequality $\mathrm{Tr}\, \rho_S^2(\tau) \leq 1$ is preserved during the evolution of the system.

Actually, the relations (49) and (51) are stronger than are needed to obtain (52). They also give an opportunity for comment on the stability of the process $z(\tau)$. Such discussion is, however, complicated by the fact that L may be a degenerate elliptic operator [30]. The degeneracy of L appears if C(z) turns out to be zero for some set N of points of R^n. The form of C(z) implies that if such a set N exists it must be a linear subspace of R^n. This is true, since $N = \bigcap_{k=-l}^{l} N(A^k)$, where $N(A^k) = \{z \in R^n : A^k z = 0\}$. Moreover, we may notice that for $z \in N$ $b(z) = 0$ and $\sigma(z) = 0$. Therefore, by (43), N is an invariant subspace with respect to the process $z(\tau)$. The decomposition of the process $z(\tau)$ for N and $N^{\perp}$ subspaces (where $N \oplus N^{\perp} = R^n$) allows us to see it as the direct sum of two vectors evolving in time. The first vector is a constant vector z_N in N, and the second is equal to the relevant diffusion process $z^{\perp}(\tau)$ in $N^{\perp}$ with strictly elliptic infinitesimal generator $L^{\perp}$ (for $z \neq 0$) (this is also clear if the equation (29) is linearly transformed so that the components in N and $N^{\perp}$ are separated).

For the generator $L^{\perp}$ it will then be possible, identically as was done above, to obtain the relation (49) (but with strong inequalities), i.e.

$$L^{\perp} v(z) < 0 \qquad \text{for} \qquad 0 < p < 1 , \quad z \neq 0. \tag{53}$$

But this is a sufficient condition for asymptotic stability in the large [29,30] of the process $z^{\perp}(\tau)$. Thus all coordinates of the process $z^{\perp}(\tau)$ converge to 0 for $\tau \to \infty$. This means that the system for $\tau \to \infty$ relaxes to the uniquely defined equilibrium position $z_{eq} = z_N \oplus O$, where $z_N \in N$ is the constant of the motion determined by initial conditions.

Let us now turn our attention to the problem of determining the subsystem evolution, i.e. to the behaviour in time of the average $Ez(\tau)$ of the process $z(\tau)$. From (43) we have immediately for $m(\tau) = E z(\tau)$

$$\frac{dm(\tau)}{d\tau} = - D\, m(\tau) , \qquad m(0) = z , \tag{54}$$

or $m(\tau) = \exp(-D\tau)z$, (55)

where D is the matrix $D = \sum_{k=-L}^{L} J_k (A^k)^{+} A^k$. The last relation gives us the rate of decay of $m(\tau)$. Thus (54) may be called the Bloch equation of the stochastic dynamical system given by (29), (1F) - (5F'), (5F"), (20), (22) and (23). The reason we choose this name becomes clear when we apply (54) to the system considered in 2.

7. Calculations for the system considered in 2

1° To find the explicit form of the limit diffusion $z(\tau)$ in the case of the example considered in this paper we must first know all matrices $\{A^k\}_{k=-2}^{2}$. Applying definitions of the basis set $\{e_i\}_{i=1}^{16}$ (12), (9), and operators $\{V^k\}_{k=-2}^{2}$ (8), (9) to (19), we get after elementary calculations the following form of matrices $\{A^k\}_{k=-2}^{2}$

$$A^{0} = \left(\frac{4}{\sqrt{6}}\right) \cdot \begin{pmatrix}
 & & & & & & & & & & & & & & & \\
 & & & & & & & & & & & 1 & & & -1 & \\
 & & & & & & & 2 & & & & & & 1 & & \\
 & & & & & & 2 & & & 1 & & & & & & \\
 & & & & & & & & & & & -1 & & & 1 & \\
 & & & & & & & & & & & & & & & \\
 & & & -2 & & & & & & & & & -1 & & & \\
 & & -2 & & & & & & & -1 & & & & & & \\
 & & & & & & & & 1 & & & & & 2 & & \\
 & & & -1 & & & & & & & & & -2 & & & \\
 & & & & & & & & & & & & & & & \\
 & -1 & & & 1 & & & & & & & & & & & \\
 & & & & & & 1 & & & 2 & & & & & & \\
 & & -1 & & & & & & -2 & & & & & & & \\
 & 1 & & & -1 & & & & & & & & & & & \\
 & & & & & & & & & & & & & & &
\end{pmatrix} \tag{56'}$$

$A^{\pm 1} = 2 \cdot$

							∓ 1						∓ 1		
							$-i$					± 1			
											i				
				∓ 1	$-i$					i					
			± 1										$-i$		
			i									i			
											∓ 1				
± 1	i									∓ 1					
														i	
														∓ 1	
			$-i$				± 1					$-i$	± 1		
		$-i$				± 1									
	∓ 1				$-i$					i					
± 1				i						∓ 1					
								$-i$	± 1						

(56'')

$A^{\mp 2} = (-2) \cdot$

											$\pm i$			$\pm i$	
											-1			-1	
												$\mp i$	1		
								$\pm i$	-1						
											-1			-1	
											$\mp i$			$\mp i$	
												1	$\pm i$		
								-1	$\mp i$						
			$\mp i$				1								
			1				$\pm i$								
$\mp i$	1			1	$\pm i$										
		$\pm i$				-1									
		-1				$\mp i$									
$\mp i$	1			1	$\pm i$										

(56''')

2° Using the explicit form of $\{A^k\}_{k=-2}^{2}$ matrices we find that the subspace $N = \{ z: A^k z = 0,\ k = 0, \pm 1, \pm 2 \}$, which is invariant with respect to the process $z(\tau)$, is equal to

$$N = x\,(e_1 + e \;\; + e_{11}) + y\, e_{16} \tag{57}$$

where x and y are arbitrary real parameters. As x and y are fixed by initial conditions ($x = x_0$ and $y = y_0$), we may write, according to our previous statements, the evolution $z(\tau)$ as

$$z(\tau) = z_N \oplus z^{\perp}(\tau) = \left[x_0 (e_1 + e_6 + e_{11}) + y_0 e_{16} \right] \oplus z^{\perp}(\tau). \tag{58}$$

From the above relation we have $Tr\, \rho_S(\tau) = Tr\, E z(\tau) = 1$ if $y_0 = \frac{1}{4}$.
Moreover, taking into account that $z^{\perp}(\tau)$ converges to 0 for $\tau \to \infty$, we obtain

$$z_{eq} = \left[x_0 (e_1 + e_6 + e_{11}) + \tfrac{1}{4} e_{16} \right] \oplus 0 \tag{59}$$

or, identifying z_{eq} with the relevant density matrix,

$$\rho_{eq} = \begin{pmatrix} \frac{1}{4}+x_0 & 0 & 0 & 0 \\ 0 & \frac{1}{4}-x_0 & 2x_0 & 0 \\ 0 & 2x_0 & \frac{1}{4}-x_0 & 0 \\ 0 & 0 & 0 & \frac{1}{4}+x_0 \end{pmatrix} \tag{60}$$

3° Applying the definition of the global average of the spin observable we may express the time evolution of z (see (25)) and ± 1 (see (26)) components of the total spin operator of our system as follows:

$$\langle S^z \rangle (\tau) = Tr\, E\left(z(\tau) \cdot \tfrac{1}{2} (S_3 \otimes S_4 + S_4 \otimes S_3) \right) = 2 \left(E z_{12}(\tau) + E z_{15}(\tau) \right) = $$
$$= 2 \left(m_{12}(\tau) + m_{15}(\tau) \right), \tag{61}$$

and

$$\langle S^{\pm 1} \rangle (\tau) = Tr\, E\left(z(\tau) \cdot \tfrac{1}{2} \left[(S_1 \otimes S_4 + S_4 \otimes S_1) \pm i (S_2 \otimes S_4 + S_4 \otimes S_2) \right] \right) =$$
$$= 2 \left[(E z_4(\tau) + E z_{13}(\tau)) \pm i (E z_8(\tau) + E z_{14}(\tau)) \right] = \tag{62}$$
$$= 2 \left[(m_4(\tau) + m_{13}(\tau)) \pm i (m_8(\tau) + m_{14}(\tau)) \right],$$

where the $m_i(\tau)$ on the right hand sides of (61) and (62) are determined by the Bloch equation (54). Let us find these elements of matrix D which are necessary to determine the explicit form of evolution for $\langle S^z \rangle$ and $\langle S^{\mp 1} \rangle$. From the definition (38) of matrix D and the form (56'), (56") and (56"') of matrices $\{A^k\}_{k=-2}^{2}$ we find that the required elements are equal to :

$$D_{12,12} = D_{15,15} = -\frac{16}{3} J_0 - 4\left(4J_1 + 8J_2\right),$$

$$D_{12,15} = D_{15,12} = \frac{16}{3} J_0 - 4\cdot 8 \cdot J_2 \,, \qquad (63)$$

$$D_{4,4} = D_{8,8} = D_{13,13} = D_{14,14} = -5\cdot\frac{16}{3} J_0 - 6\cdot 4\cdot J_1 - 4\cdot 4\cdot J_2 \,,$$

$$D_{4,13} = D_{13,4} = D_{8,14} = D_{14,8} = -4\cdot\frac{16}{6} J_0 - 4\cdot 4\cdot J_1 \,,$$

and $D_{12,m} = D_{15,m} = D_{4,m} = D_{8,m} = D_{13,m} = D_{14,m} = 0$ for any other m than considered above.

From these values of D_{ij}, $i,j = 1, \ldots, 16$, and the Bloch equation (54), it follows that

$$\frac{d}{d\tau} \langle S^z \rangle(\tau) = -4\left(4J_1 + 16J_2\right)\langle S^z \rangle(\tau)\,; \quad \langle S^z \rangle(0) = m_0^z \qquad (64)$$

and $$\frac{d}{d\tau} \langle S^{\pm 1} \rangle(\tau) = -4\left(6J_0 + 10J_1 + 4J_2\right)\langle S^{\pm 1} \rangle(\tau)\,, \quad \langle S^{\pm 1} \rangle(0) = m_0^{\pm 1}. \qquad (65)$$

The above two equations give

$$\langle S^z \rangle(\tau) = m_0^z \cdot \exp\left(-\frac{\tau}{T_1}\right) \qquad (66)$$

and $$\langle S^{\pm 1} \rangle(\tau) = m_0^{\pm 1} \cdot \exp\left(-\frac{\tau}{T_2}\right), \qquad (67)$$

where $$\frac{1}{T_1} = 4\left(4J_1 + 16J_2\right), \qquad (68)$$

and $$\frac{1}{T_2} = 4\left(6J_0 + 10J_1 + 4J_2\right). \qquad (69)$$

Here T_1 is to be interpreted as the relaxation time of the z - component of the total spin operator (longitudinal relaxation time), and T_2 is to be interpreted as the relaxation time of the perpendicular to the z - component of the total spin operator (perpendicular relaxation time).

Acknowledgements

The author wishes to thank Professor J.T. Lewis for valuable suggestions concerning the problem here discussed, Professor J.R. McConnell and Professor N.G. Van Kampen for helpful comments and B. Lenoach for enlightening discussions on applications of asymptotic analysis of stochastic equations.

References

1. Kubo, R., Lectures in Theoretical Physics I, ed. by Brittin, W.E., and Dunham, L.G., Interscience Publishers, New York, 1959.
2. Kubo, R., Fluctuation, Relaxation and Resonance in Magnetic Systems, ed. by ter Haar, Oliver and Boyd, Edinburgh, 1962.
3. Kubo, R., J. Math. Phys. 4, 174 (1963).
4. Van Kampen, N.G., Physica 74, 215 (1974).
5. Van Kampen, N.G., Phys. Rep. 24C, 172 (1976).
6. Redfield, A.G., IBM J. Research Develop. 1, 19 (1957).
7. Howe, H.S., J. Fluid Mech. 45, 769 (1971).
8. Bourret, R.C., Can. J. Phys. 40, 782 (1962).
9. Bourret, R.C., Nuovo Cim. 26, 1 (1962).
10. Keller, J.B., Proc. Symp. Appl. Math. 13, 227 (1962).
11. Van Hove, L., Physica 21, 517 (1955).
12. Van Hove, L., Physica 22, 342 (1956).
13. Stratonovich, R.L., Conditional Markov Processes and their Application to the Theory of Optimal Control, Elsvier, New York, 1968.
14. Khasminskii, R.Z., Theory Prob. Appl. 11, 390 (1966).
15. Papanicolaou, G.C., Varadhan, S.R.S., Comm. Pure Appl. Math. 26, 497 (1973).
16. Papanicolaou, G.C., Kohler, W., Comm. Pure Appl. Math., 27, 641 (1974).
17. Ventzel, A.D., Freidlin, M.J., Fluktuacii v Dinamitcheskikh Sistiemakh pod Deistviem Malykh Slutchainykh Vozmushtchenii, Nauka, Moskva, 1979.
18. Tomita, K., Prog. Theor. Phys. (Kyoto) 19, 541 (1958).
19. Abragam, A., The Principles of Nuclear Magnetism, Clarendon Press, Oxford, 1961.
20. Hubbard, P.S., Rev. Mod. Phys. 33, 249 (1961).
21. Spohn, M., Rev. Mod. Phys. 53, 569 (1980).
22. Blombergen, N., Purcell, E.M., Pound, R.V., Phys. Rev. 73, 679 (1948).
23. Ford, G.W., Lewis, J.T., McConnell, J.R. Phys. Rev. A 19, 907 (1979).
24. Hubbard, P.S., Phys. Rev. 180, 319 (1969).

25. McConnell, J.R. Rotational Brownian Motion and Dielectric Theory, Academic Press, London, 1980.
26. Rozanov, Y.A., Stationary Random Processes, San Francisco, 1967.
27. Ibragimov, I.A., Rozanov, Y.A., Gaussian Random Processes, Springer-Verlag, New York, 1978.
28. Khasminskii, R.Z., Theory Prob. Appl. 11, 211 (1966).
29. Arnold, L., Stochastic Differential Equations, John Wiley & Sons, New York, 1974.
30. Friedman, A., Stochastic Differential Equations and Applications, Academic Press, New York, 1975.