

Brownian Motion of Spins

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Stochastic models of Brownian motion of a spin are discussed. If a frictional resistance is assumed to accompany the random field causing Brownian motion, the stochastic equation of spin motion leads to a Fokker-Planck equation which guarantees approach to thermal equilibrium. With a Landau-Lifshitz type friction assumed, the average magnetic moment is shown to obey the Bloch equation.

§1. Introduction

In a condensed system the motion of a spin is very much complicated by the influence of interaction with its surroundings. The complexity itself allows, however, an idealization, namely introduction of a stochastic model. Here we consider, as the simplest idealization, a Brownian motion model, in which the interaction of a spin with the surroundings is represented by a randomly fluctuating magnetic field. Then the motion of a spin would be described by a stochastic equation of the type

$$\frac{d\mathbf{M}}{dt} = \gamma \{ \mathbf{H}(t) + \mathbf{H}'(t) \} \times \mathbf{M}, \quad (1)$$

where $\mathbf{H}(t)$ is the external field controlled in the process of observation and $\mathbf{H}'(t)$ is a stochastic magnetic field. Some years ago, one of the authors discussed briefly such a model of a classical spin on the basis of his theory of the stochastic Liouville equation.²⁾ Yoshimori and Korringa³⁾ developed a similar theory based on a stochastic Heisenberg equation of a quantal spin. When the fluctuating field is assumed to be a Gaussian process and to satisfy the narrowing condition,¹⁾ these theories give the Bloch equation for the magnetization. There is, however, a flaw in the equation thus derived. Namely, the magnetization does not relax towards its equilibrium value corresponding to the given external field but towards the zero value. We cannot incorporate in the theory a finite temperature and the corresponding equilibrium magnetization.

In order to overcome this difficulty some attempts have been made. Korringa⁴⁾ introduced a complex time into a stochastic Schrödinger equation. Its imaginary part corresponds to the reciprocal temperature. Using this formalism,

Korringa et al.⁵⁾ obtained a modified Bloch equation, by which the magnetization relaxes to a finite equilibrium value. Nakano and Yoshimori⁶⁾ made another approach with the aid of a path-probability method developed by Kikuchi.⁷⁾ Essentially they assume a Chapman-Smoluchowski equation (a master equation) of the type

$$\frac{dp_i}{dt} = \sum_j \theta_{ij} (p_j e^{(E_j - E_i)/2kT} - p_i e^{(E_i - E_j)/2kT}),$$

where i and j refer to the states of the system (spin) in consideration, p_j the probability to find the system in the j -th state and E_j is the energy of the j -th state. They used the stochastic equation of motion of the type (1) for determining the transition probability θ_{ij} . They introduced, however, rather arbitrarily the factors $\exp(\pm(E_i - E_j)/2kT)$ in order to assure the approach to equilibrium at a finite temperature.

Here we consider the problem starting from a somewhat different point of view that corresponds to the Langevin theory of Brownian motion. As is well known in the theory of Brownian motion the random force acting on a Brownian particle is necessarily combined with a frictional force, a fact which represents a very general law of nature—the fluctuation dissipation theorem.⁸⁾ This means that the stochastic equation of motion, Eq. (1), must be supplemented with a frictional force. Equation (1) then will be replaced by such an equation as

$$\frac{d\mathbf{M}}{dt} = \gamma \left\{ \mathbf{H}(t) + \mathbf{H}'(t) - \kappa \frac{d\mathbf{M}}{dt} \right\} \times \mathbf{M}. \quad (2)$$

The frictional force is related in some way to the random field \mathbf{H}' by the fluctuation dissipation theorem.

Compared to the familiar theory of Brownian motion of a free particle or a harmonic oscillator, the Brownian motion of spin involves some complexities. These complexities come from the quasi-nonlinear structure of Eq. (1) or (2). The magnetization $\mathbf{M}(t)$ is not linear in the stochastic field $\mathbf{H}'(t)$, so that a simple harmonic analysis cannot be employed. In fact, for a general case, either Eq. (1) or (2) is hard to solve even if the basic process $\mathbf{H}'(t)$ is assumed to be simple, for example, a Gaussian process. The stochastic process of $\mathbf{M}(t)$ is then rather complicated, not allowing easily an analytical treatment. Some attempts have been made⁹⁾¹⁰⁾ for such a problem with the purpose to apply it to the magnetic resonance at low fields.

In this paper we shall not, however, go into such a complicated problem, but we confine to the narrowing limit case, where the random field is weak and fluctuating very fast so that the condition

$$\Delta\tau_c \ll 1, \quad \Delta \simeq \gamma H' \quad (3)$$

is satisfied, where H' is the average amplitude, and τ_c the correlation time of

the random field. In this limit, the stochastic process of $\mathbf{M}(t)$ is described by a Fokker-Planck equation. On the average the magnetization vector will be shown to obey the Bloch equation with relaxation to a finite thermal equilibrium.¹¹⁾

§2. A classical spin rotating without friction

In order to make the nature of the problem clear, we start here with the simplest stochastic model of fluctuating spin motion as is represented by Eq. (1). It is most convenient for treating this type of stochastic process to introduce a stochastic Liouville equation²⁾ which is written in this case as

$$\frac{\partial}{\partial t} \rho(\mathbf{M}, t) = -\frac{\partial}{\partial \mathbf{M}} \cdot (\dot{\mathbf{M}} \rho(\mathbf{M}, t)) \quad (4)$$

or

$$\frac{\partial}{\partial t} \rho = -i \mathcal{L}(t) \rho, \quad (5)$$

where the Liouville operator \mathcal{L} is written as

$$\mathcal{L} = \mathcal{L}_0 + \mathcal{L}'(t) \quad (6)$$

with

$$\mathcal{L}_0 = \omega_0 L_z, \quad (7)$$

$$\mathcal{L}'(t) = \gamma \mathbf{H}'(t) \cdot \mathbf{L}, \quad (8)$$

$$\mathbf{L} = -i \mathbf{M} \times \frac{\partial}{\partial \mathbf{M}}. \quad (9)$$

The direction of \mathbf{H}_0 is taken as the z -axis and $\omega_0 = \gamma H_0$ is the Larmor frequency of the spin in \mathbf{H}_0 . The operator \mathbf{L} is an angular momentum operator in the space of \mathbf{M} . Equation (4) or (5) is a stochastic Liouville equation for the probability density $\rho(\mathbf{M}, t)$ to find the vector \mathbf{M} at the time t in the neighbourhood of a given point in the space of \mathbf{M} . It contains the stochastic process $\mathbf{H}'(t)$ from which the stochastic properties of $\mathbf{M}(t)$ should be determined. Equation (5) can be formally solved for an initial distribution $\rho(\mathbf{M}, 0)$ and for a given sample of process $\mathbf{H}'(t)$ as

$$\rho(\mathbf{M}, t) = \exp_{\leftarrow} \left\{ -i \int_0^t \mathcal{L}(t') dt' \right\} \rho(\mathbf{M}, 0). \quad (10)$$

In particular, for

$$\rho(\mathbf{M}, 0) = \delta(\mathbf{M} - \mathbf{M}')$$

the final distribution can be written as

$$\rho(\mathbf{M}, t) = (\mathbf{M} | \exp_{\leftarrow} \{ -i \int_0^t \mathcal{L}(t') dt' \} | \mathbf{M}'), \quad (11)$$

where the bra-ket notation is used to represent the integral kernel of the transformation. This transformation is averaged over the whole ensemble of $\mathbf{H}'(t)$ to give the transition probability from \mathbf{M}' to \mathbf{M} in the time interval $(0, t)$,

$$f(\mathbf{M}t | \mathbf{M}'0) = (\mathbf{M} | \langle \exp \{ -i \int_0^t \mathcal{L}(t') dt' \} \rangle | \mathbf{M}'), \quad (12)$$

where $\langle \rangle$ means the ensemble average. Thus we may call

$$\phi(t) = \langle \exp \{ -i \int_0^t \mathcal{L}(t') dt' \} \rangle \quad (13)$$

the transition operator of the process $\mathbf{M}(t)$. The problem now is to calculate this transition operator (13) and to find averages of relevant physical quantities.

This is not an easy task. Generally it is not possible to derive an analytic expression of the transition probability unless some simplification is introduced. It may be reasonably assumed that the process $\mathbf{H}'(t)$ is Gaussian. The Brownian spin motion under this assumption has been treated in a greater detail by Toyabe¹⁰⁾ and Toyabe and Kubo.⁹⁾ Even with this simplification no exact treatment is possible because the operator $\mathcal{L}(t)$ for different time points are not commutable. Only in the limit of narrowing condition (3), the theory becomes simple. If, however, the narrowing condition (3) is satisfied, the same simplification can be achieved not only for a Gaussian modulation $\mathbf{H}'(t)$ but also for a more general class of the process $\mathbf{H}'(t)$ as long as it is sufficiently moderate. We do not try here to give a rigorous definition of a moderate process, but we mean by this a process for which a certain kind of central limit theorem holds. Thus a process consisting of a series of irregular pulses is excluded.

In the interaction representation defined by

$$\hat{\rho} = e^{i\mathcal{L}_0 t} \rho$$

and

$$\mathcal{Q}(t) = e^{i\mathcal{L}_0 t} \mathcal{L}'(t) e^{-i\mathcal{L}_0 t} \quad (14)$$

the transition operator (13) is written as

$$\phi(t) = e^{-i\mathcal{L}_0 t} \langle \exp \{ -i \int_0^t \mathcal{Q}(t') dt' \} \rangle e^{i\mathcal{L}_0 t}, \quad (15)$$

where only the operator $\mathcal{Q}(t)$ contains the random field $\mathbf{H}'(t)$ for which we assume that

$$\langle \mathbf{H}'(t) \rangle = 0,$$

$$\langle H'_\alpha(t_1) H'_\beta(t_2) \rangle = 0, \quad \alpha \neq \beta, \quad (16)$$

$$\gamma^2 \langle H'_x(t_1) H'_x(t_2) \rangle = \gamma^2 \langle H'_y(t_1) H'_y(t_2) \rangle = \psi_{\mathbf{H}}(t_1 - t_2), \quad (17)$$

$$r^2 \langle H'_z(t_1) H'_z(t_2) \rangle = \psi_{\parallel}(t_1 - t_2). \quad (18)$$

When the ordered exponential operator on the right hand side of Eq. (15) is expanded, the first term becomes

$$\begin{aligned} & - \int_0^t dt_1 \int_0^{t_1} dt_2 e^{i\mathcal{L}_0 t_1} \langle \mathcal{L}'(t_1) e^{-i\mathcal{L}_0(t_1-t_2)} \mathcal{L}'(t_2) e^{i\mathcal{L}_0(t_1-t_2)} \rangle e^{-i\mathcal{L}_0 t_1} \\ & \simeq - \int_0^t dt_1 e^{i\mathcal{L}_0 t_1} \int_0^\infty \langle \mathcal{L}'(t_1) e^{-i\mathcal{L}_0 \tau} \mathcal{L}'(t_1 - \tau) e^{i\mathcal{L}_0 \tau} \rangle e^{-i\mathcal{L}_0 t_1} \\ & = - \int_0^t dt_1 e^{i\mathcal{L}_0 t_1} \left\{ i\delta\omega_0 L_z + \frac{1}{2\tau_1} (L_x^2 + L_y^2) + \frac{1}{2\tau_0} L_z^2 \right\} e^{-i\mathcal{L}_0 t_1}, \end{aligned} \quad (19)$$

where

$$\begin{aligned} \frac{1}{2\tau_0} &= \int_0^\infty \psi_{\parallel}(t) dt, \\ \frac{1}{2\tau_1} &= \int_0^\infty \psi_{\perp}(t) \cos \omega_0 t dt, \\ \delta\omega_0 &= \int_0^\infty \psi_{\perp}(t) \sin \omega_0 t dt. \end{aligned} \quad (20)$$

In transforming Eq. (19) we used the assumption that the correlation time τ_c of ψ_{\parallel} or ψ_{\perp} is very short compared to the time t under consideration, namely

$$t \gg \tau_c, \quad (21)$$

thus allowing to extend the lower limit of t_2 to $-\infty$. Higher order expansion terms of averaged ordered exponential operator in Eq. (15) can be evaluated in a similar manner. As long as the random field $\mathbf{H}'(t)$ is moderate and satisfies the narrowing condition (3), only important terms are those which are reduced to products of the operator (19) so that the transition operator (15) has the form

$$\begin{aligned} \Phi(t) &= e^{-i\mathcal{L}_0 t} \exp \left[- \int_0^t dt_1 e^{i\mathcal{L}_0 t_1} \left\{ i\delta\omega_0 L_z + \frac{1}{2\tau_1} (L_x^2 + L_y^2) \right. \right. \\ & \quad \left. \left. + \frac{1}{2\tau_0} L_z^2 \right\} e^{-i\mathcal{L}_0 t_1} \right] e^{i\mathcal{L}_0 t}. \end{aligned}$$

By differentiating the above expression with respect to t , we find

$$\frac{\partial}{\partial t} \Phi(t) = -i(\omega_0 + \delta\omega_0) L_z + \frac{1}{2\tau_1} (L_x^2 + L_y^2) + \frac{1}{2\tau_0} L_z^2. \quad (22)$$

Thus the transition probability f is the fundamental solution of the equation,

$$\left[\frac{\partial}{\partial t} + i(\omega_0 + \delta\omega_0) L_z + \frac{1}{2\tau_1} (L_x^2 + L_y^2) + \frac{1}{2\tau_0} L_z^2 \right] f = 0. \quad (23)$$

This may be written as

$$\left[\frac{\partial}{\partial t} + i\gamma(1+\delta)\mathbf{H}_0 \cdot \mathbf{L} - (i\mathbf{L} \cdot \mathbf{D} \cdot i\mathbf{L}) \right] f = 0, \quad (24)$$

where \mathbf{H}_0 is the external field and the diffusion tensor \mathbf{D} is defined by

$$\mathbf{D} = D_1 \left(1 - \frac{\mathbf{H}_0 \mathbf{H}_0}{H_0^2} \right) + D_0 \frac{\mathbf{H}_0 \mathbf{H}_0}{H_0^2}, \quad D_1 = \frac{1}{2\tau_1}, \quad D_0 = \frac{1}{2\tau_0}. \quad (25)$$

The relaxation times τ_0 and τ_1 and the fractional shift $\delta = \delta\omega_0/\omega_0$ are defined by Eq. (20). Equation (24) can be used for a time-dependent external field $\mathbf{H}_0(t)$ as long as its time variation is slow in the correlation time τ_c .

The average of the magnetic moment

$$\langle \mathbf{M}(t) \rangle = \int \mathbf{M} f(\mathbf{M}, t) d\mathbf{M}$$

is easily found to satisfy the Bloch equation

$$\begin{aligned} \frac{d}{dt} \langle M_x \rangle &= -\omega'_0 \langle M_y \rangle - \frac{\langle M_x \rangle}{T_2}, & \omega'_0 &= \omega_0 + \delta\omega_0, \\ \frac{d}{dt} \langle M_y \rangle &= \omega'_0 \langle M_x \rangle - \frac{\langle M_y \rangle}{T_2}, \\ \frac{d\langle M_z \rangle}{dt} &= \frac{\langle M_z \rangle}{T_1}, \end{aligned} \quad (26)$$

where the longitudinal and the transverse relaxation times T_1 and T_2 are given by

$$T_1 = \tau_1, \quad \frac{1}{T_2} = \frac{1}{2} \left(\frac{1}{\tau_0} + \frac{1}{\tau_1} \right). \quad (27)$$

By Eq. (26) the magnetic moment relaxes to zero. As was mentioned in Introduction, this necessarily follows from Eq. (1) which lacks the friction term.

§3. A classical spin rotating with friction

We now consider a classical spin which is subject to a frictional resistance as it rotates. Following Landau and Lifshitz¹²⁾ we may assume the equation of motion,

$$\frac{d}{dt} \mathbf{M} = \gamma \mathbf{H} \times \mathbf{M} - \eta (\mathbf{H} \times \mathbf{M}) \times \mathbf{M}. \quad (28)$$

This is obtained from the observation that $d\mathbf{M}/dt$ is a linear combination of $\mathbf{H} \times \mathbf{M}$ and $(\mathbf{H} \times \mathbf{M}) \times \mathbf{M}$ if the magnitude of \mathbf{M} should be represerved. We may equally well assume instead another type of equation,

$$\frac{d\mathbf{M}}{dt} = \gamma' \left(\mathbf{H} - \kappa \frac{d\mathbf{M}}{dt} \right) \times \mathbf{M} \quad (29)$$

as was mentioned in Introduction. The latter equation can be transformed into

$$\frac{d\mathbf{M}}{dt} = \frac{\gamma'}{1 + (\gamma'\kappa)^2 M^2} (\mathbf{H} - \kappa \mathbf{H} \times \mathbf{M}) \times \mathbf{M}$$

which is same as Eq. (28) only with renormalized coefficients. Thus we use here Eq. (28).

If the field \mathbf{H} consists of the external field $\mathbf{H}_0(t)$ and the random field $\mathbf{H}'(t)$, namely

$$\mathbf{H}(t) = \mathbf{H}_0(t) + \mathbf{H}'(t),$$

Eq. (28) is our stochastic equation of motion for a classical spin. This may further be simplified to

$$\frac{d}{dt} \mathbf{M}(t) = \gamma (\mathbf{H}_0(t) + \mathbf{H}'(t)) \times \mathbf{M} - \eta (\mathbf{H}_0 \times \mathbf{M}) \times \mathbf{M} \quad (30)$$

by omission of the random field in the friction term. As we shall see later, the Einstein relation requires that

$$\eta = 1/2\tau_1 kT, \quad (31)$$

so we have the order of magnitude estimation,

$$O(\eta \mathbf{H}' \times \mathbf{M}) = H'M/2\tau_1 kT = \Delta^2 \tau_e H'M/kT. \quad (32)$$

Because of the narrowing condition and because of the usual experimental condition that

$$H'M \ll kT,$$

the effect of random part of the friction term is much smaller than the direct effect of the random field, so that the above simplification is justified.

Derivation of the Fokker-Planck equation from (31) is straightforward under the assumptions of coarse graining (21), narrowing (3) and the moderate behavior of $\mathbf{H}'(t)$. The unperturbed Liouville operator, Eq. (7), is now replaced by

$$\mathcal{L}_0(t) = \omega_0 L_z + \eta H_0 (L_x M_y - L_y M_x). \quad (33)$$

The presence of the second term on the right hand side of Eq. (33) may seem to introduce complications. However, the same estimation as (32) shows that it causes only very slow time change of $\exp(i\mathcal{L}_0 t)$ in comparison with the time rate characterized by the correlation time τ_e of the random field, so that the operator in the bracket in Eq. (19) need not be changed. Thus the only change to Eq. (24) is addition of the term

$$\frac{\partial}{\partial \mathbf{M}} \eta (\mathbf{H}_0 \times \mathbf{M}) \times \mathbf{M} f = i\eta \mathbf{L}(\mathbf{M} \times \mathbf{H}_0) f.$$

So Eq. (24) is now generalized to

$$\left[\frac{\partial}{\partial t} + i\gamma(1+\delta)\mathbf{H}_0\mathbf{L} + i\mathbf{LD}\left(\mathbf{L} - \frac{\mathbf{M} \times \mathbf{H}_0}{kT}\right) \right] f = 0 \quad (34)$$

which includes the effect of friction associated with the random field. Here we have already used the relation (31). In the polar coordinates defined by

$$M_x = M \sin \theta \cos \phi, \quad M_y = M \sin \theta \sin \phi, \quad M_z = M \cos \theta,$$

Eq. (35) is written as

$$\begin{aligned} \left(\frac{\partial}{\partial t} + \omega'_0 \frac{\partial}{\partial \phi} \right) f = & \left[\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left\{ \sin \theta D_1 \left(\frac{\partial}{\partial \theta} + \frac{H_0 M \sin \theta}{kT} \right) \right\} \right. \\ & \left. + \frac{1}{\sin^2 \theta} \frac{\partial}{\partial \phi} \left(D_0 \frac{\partial}{\partial \phi} \right) \right] f \end{aligned} \quad (35)$$

which shows that the function

$$\exp(H_0 M \cos \theta / kT)$$

is in fact the equilibrium distribution in a static field. If further the extreme narrowing condition

$$\omega_0 \tau_c \ll 1$$

is satisfied, the relaxation times τ_0 and τ_1 become equal, the diffusion tensor D becomes isotropic, and Eq. (35) becomes just the rotational diffusion equation of an electric dipole emersed in a viscous liquid, as was shown many years ago by Debye.¹³⁾

The equation of motion for the averaged magnetic moment $\langle M \rangle$ is easily derived from Eq. (34) and is given by

$$\begin{aligned} \frac{d\langle M_x \rangle}{dt} &= -\omega'_0 \langle M_y \rangle - \frac{1}{T_2} \langle M_x \rangle - \frac{H_0 \langle M_x M_z \rangle}{2 T_1 k T}, \\ \frac{d\langle M_y \rangle}{dt} &= \omega'_0 \langle M_x \rangle - \frac{1}{T_2} \langle M_y \rangle - \frac{H_0 \langle M_y M_z \rangle}{2 T_1 k T}, \\ \frac{d\langle M_z \rangle}{dt} &= -\frac{1}{T_1} \langle M_z \rangle + \frac{H_0 \langle M_x^2 + M_y^2 \rangle}{2 T_1 k T}. \end{aligned} \quad (36)$$

Due to the non-linearity of the basic equation (30), these equations include averages of higher moments and make in general a hierarchy of equations. If deviation from equilibrium is small, this hierarchy may be decoupled. In equilibrium the third equation of Eqs. (36) gives

$$\langle M_z \rangle = H_0 \langle M_x^2 + M_y^2 \rangle / 2kT = H_0 \langle M_z^2 \rangle / kT. \quad (37)$$

Equation (37) shows that the magnetic susceptibility χ is given by

$$\chi = \langle M_x^2 \rangle_0 / kT \quad (38)$$

in accordance with the well-known result of statistical mechanics.

As long as the external field is not too strong so that $H_0 M \ll kT$, the last terms on the right hand sides of the first and second equations of (36) are negligible as damping to $\langle M_x \rangle$ and $\langle M_y \rangle$, and so we have the Bloch equations

$$\begin{aligned} \frac{d}{dt} \langle M_x \rangle &= -\omega'_0 \langle M_y \rangle - \frac{1}{T_2} \langle M_x \rangle, \\ \frac{d}{dt} \langle M_y \rangle &= \omega'_0 \langle M_x \rangle - \frac{1}{T_2} \langle M_y \rangle, \\ \frac{d}{dt} \langle M_z \rangle &= -\frac{1}{T_1} (\langle M_z \rangle - \chi H_0). \end{aligned} \quad (39)$$

It is interesting to note that thus we have been led to the Bloch-type relaxation in spite of the fact that we started from the Landau-Lifshitz friction in Eq. (28).

Equation (34) contains relaxation of the average magnetic moment towards the equilibrium value corresponding to the instantaneous magnetic field. This equation may be used for studying the response and the noise of magnetic spins when the external field $H_0(t)$ is changing in time, provided that it varies in time slowly enough in comparison with the fluctuating field.

§4. Concluding remarks

We have shown that a phenomenological theory can be formulated for a classical spin under the influence of a random field which represents the interaction with the surroundings. If the random field is assumed to be weak, moderate, and sufficiently short-correlated, it is possible to derive a Fokker-Planck equation for the stochastic motion of the spin moment. With a friction force accompanying the random field, the Fokker-Planck equation assures the approach to equilibrium at a finite temperature. The extension of this treatment to a quantal spin seems easy from a formal point of view. In quantum mechanics we can no longer determine all the components of a spin so that we are obliged to think in terms of the density matrix, the equation of motion of which is given by

$$\frac{\partial \rho}{\partial t} = \frac{1}{i\hbar} [\mathcal{H}(t), \rho]. \quad (40)$$

The density matrix of a spin can be expanded in the operator spherical harmonics, the first few members of which are

$$\begin{aligned} &1, S_x, S_y, S_z, S_x S_y + S_y S_x, S_y S_z + S_z S_y, S_z S_x + S_x S_z, \\ &S_x^2 - S_y^2, 2S_z^2 - S_x^2 - S_y^2, \dots \end{aligned}$$

Thus we may put

$$\rho = A + c\mathbf{M}(t)\mathbf{S} + \dots, \quad (41)$$

where the expansion coefficient \mathbf{M} is the quantum mechanical expectation of \mathbf{S} at the time t , namely

$$\mathbf{M}(t) = \text{Tr} \rho(t) \mathbf{S}, \quad (42)$$

is the normalization is so chosen that

$$c^{-1} = \text{Tr} S_x^2 = (2S+1)S(S+1).$$

If the Hamiltonian $\mathcal{H}(t)$ is

$$\mathcal{H}(t) = (\mathbf{H}_0 + \mathbf{H}') \gamma \hbar \mathbf{S}$$

Eq. (40) gives the equation

$$\frac{d\mathbf{M}}{dt} = \gamma(\mathbf{H}_0 + \mathbf{H}') \times \mathbf{M}$$

which is the stochastic equation of motion for $\mathbf{M}(t)$, or the quantum-mechanical expectation of \mathbf{S} . From a phenomenological point of view this equation may again be supplemented with a friction term in order to guarantee the approach to thermal equilibrium. Then the treatments discussed in the preceding sections may be regarded to apply to a quantal spin as well as to a classical spin. An average over the distribution of \mathbf{M} means an average of a quantum-mechanical expectation over the ensemble of the stochastic process of the random field $\mathbf{H}'(t)$. There remains, however, certain ambiguities concerning the meaning of physical observations. For example, it is not very clear how to interpret the correlation function of the type $\langle \mathbf{M}(t_1) \mathbf{M}(t_2) \rangle$. Generally speaking the Brownian motion theory of a quantum system is at present rather at an unsatisfactory state and remains an outstanding problem in statistical mechanics. Here we shall not go any further into this conceptually complex problem.

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