

# Nonequilibrium properties of structural and spin glasses in the quantum case

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The molecular field approximation and the Keldysh technique are used to develop a general theory of nonlinear nonequilibrium processes in structural and spin glasses in the quantum case. A calculation is reported of a nonequilibrium single-particle distribution function of molecular fields. A description is given of slow relaxation of the profiles of lines in the optical spectra of structural order-disorder glasses. The three-particle correlator in a Heisenberg spin glass is calculated.

## 1. INTRODUCTION

The molecular-field approximation is used in the present paper to develop a general theory of nonlinear nonequilibrium processes in quantum glasses. For brevity, we shall use the single term “structural glasses” for dipole and proton glasses and we shall also consider jointly spin glasses. From the physical point of view these three types of glass differ only in respect of the specific nature of the Hamiltonian, which is not very important in the general theory. Therefore, all the systems discussed below will be simply called quantum glasses, bearing in mind that in all cases the quantum effects are important. In discussing specific results we shall naturally employ specific Hamiltonians.

There is a highly developed theory of the molecular field for spin glasses in the classical limit both for the static case and for the dynamic equilibrium case (see, for example, the reviews in Refs. 1 and 2). However, nonequilibrium processes, which form the basis of all the physics of the investigated systems, have been largely ignored (see, for example, Refs. 1, 3, and 18). There is as yet no general theory of these processes for macroscopic times. The present paper is specifically intended to provide such a theory. We shall consider the most general case of quantum properties because they are important in the case of both structural and spin glasses.

The following comments should be made. Unfortunately, it is very difficult to go beyond the molecular-field approximation. Therefore, a natural question arises whether the results obtained in this approximation represent reality. It is generally accepted<sup>1-3</sup> that the molecular field theory describes experiments qualitatively and can serve as the language for discussing the phenomena observed experimentally. However, no attempt should be made to carry out any specific quantitative fitting of the theory to the experimental results and only the qualitative approach should be employed. We hope that this applies also to the description of nonequilibrium processes in quantum glasses for the full range of times (from quantum to macroscopic), governing slow relaxation, although this can naturally be demonstrated only by going outside the molecular-field approximation.

In an earlier communication<sup>4</sup> the present author developed a static theory of quantum glasses under nonergodic conditions. This problem has been known for a long time,<sup>5</sup> but it was solved first only recently.<sup>4</sup> It was demonstrated there that nonergodic and quantum properties can be separated in a certain sense and that a closed system of equations

can be written down for all the quantities of interest to us.

This theory includes naturally the Matsubara times  $\tau$  and the Matsubara frequencies  $\omega_n = 2\pi nT$ . It would seem that a description of any irreversible process, including one which is strongly nonlinear, can be obtained by analytic continuation using appropriate Matsubara frequencies. However, it is found that this approach can give only the intravalley susceptibilities. Intervalley transitions simply do not appear when analytic continuation is employed.

This is naturally due to nonergodicity of our systems. A theorem on analytic continuation has been derived for ergodic systems and may not be valid in the nonergodic case. Physically this is due to the fact that in developing a static theory it is assumed implicitly that in the limit  $t = -\infty$  (where  $t$  is time) a Gibbs distribution is already established. Some years ago, on the other hand, the problem of the role of the initial conditions in the physics of spin glasses has been considered.<sup>6-8</sup> It has been found that the initial conditions play a decisive role in these systems. If we assume that a Gibbs distribution exists for  $t = -\infty$ , then only intravalley dynamics is obtained and nothing else. This is a classical analog of our conclusion in the quantum case.

However, if this assumption is not made a completely different solution is clearly obtained. The authors of the cited investigations have been unable to identify the nature of this solution or its physical meaning. They have simply postulated that this is the Sompolinsky solution<sup>9-11</sup> closely related to the problems investigated by that author in the steady-state dynamic case,<sup>1,3,12,13</sup> but this could not be proved in Refs. 6–8.

However, it is clear from the results of Refs. 6–8 that in investigating the steady-state dynamic solution with intervalley dynamics or in study of transient phenomena we cannot begin with a Gibbs initial state, but we must assume that the zeroth state at  $t = -\infty$  is of different nature.

We shall use the Keldysh technique.<sup>14</sup> In this case a random exchange interaction is included either to the zeroth Hamiltonian or in the interaction form. We can easily show (we shall not do this, but the reader is directed to, for example, Ref. 15) that in the former case it is necessary to use, exactly as in the static case, the replica method and this yields an analytic continuation of the solution obtained by the present author.<sup>4</sup> This is to be expected if we bear in mind that a Gibbs distribution is assumed to exist at  $t = -\infty$ . In the second case it is found that there is no need to use any

replicas, but it is essential to assume the existence of slow logarithmic dynamics and to attribute the nonergodicity to breakdown of the fluctuation–dissipation theory.

This slow dynamics corresponding to intervalley transitions is in the steady-state case fully analogous to that discussed by the present author and Horner.<sup>1,3,13,16–18</sup>

However, in the transient case, which is of main interest to us, we shall construct a general theory of nonlinear non-equilibrium processes in quantum glasses.

It is very interesting to consider how one can avoid the use of the replica method. It is found that the Keldysh technique includes a perfectly general mechanism which causes vacuum loops to vanish. It is this mechanism that replaces the replica method.

After developing a general theory, we shall consider a number of specific tasks and calculate the corresponding generalized nonlinear susceptibilities. We shall consider only two problems. In the earlier paper<sup>4</sup> it was pointed out that there is a problem of slow relaxation of the spectrum of structural glasses on application of an external field. We shall solve this problem below to arbitrary order in terms of an external field, i.e., we shall tackle the general nonlinear problem.

The second task is related to a three-particle correlation function of a Heisenberg spin glass. It is well known that at finite frequencies a three-frequency correlation function of a Heisenberg ferromagnet differs from zero even in the paramagnetic range of temperatures. This gives rise to a number of interesting phenomena in the scattering of polarized light or of polarized neutrons. We shall study such a correlation function for a spin glass and also consider the problem of its slow relaxation under nonlinear conditions.

## 2. DERIVATION OF THE PRINCIPAL EQUATIONS

We shall consider, as stated in the Introduction, Hamiltonians of three types. The first is the Hamiltonian of dispersive structural glasses (which include dipole glasses):

$$H = - \sum_{ik} J_{ik} m_i m_k + \sum_i U(m_i, r_i),$$

$$U(m, r) = m^2/2b + um^4/8 + b\omega_0^2 r^2/2, \quad (1)$$

where  $m$  and  $r$  are the displacement and momentum, made dimensionless as a result of division by the lattice constant;  $\omega_0$  is the oscillator frequency. This Hamiltonian was discussed in detail in Ref. 4.

The second Hamiltonian represents order–disorder structural glasses<sup>4</sup>

$$H = - \sum_{ik} J_{ik} S_i^z S_k^z - \Delta \sum_i S_i^x, \quad (2)$$

where  $S_i^z$  and  $S_i^x$  are the spin operators for the spin  $S = 1/2$ , whereas  $\Delta$  is a constant governing tunneling.

The third system describes ordinary Heisenberg spin glasses with the Hamiltonian

$$H = - \sum_{ik} J_{ik} S_i S_k - h \sum_i S_i^z, \quad (3)$$

where  $h$  is the external magnetic field. In all cases the quantity  $J_{ik}$  is the random exchange interaction with the average value amounting to zero and with

$$\langle J_{ik}^2 \rangle = I_{ik}. \quad (4)$$

We shall write down the general expressions for the Hamiltonian of Eq. (1), and the generalization to the other Hamiltonians will be self-evident.

The zeroth Hamiltonian in Eq. (1) will be assumed to be the quadratic part of  $U(m, r)$ :

$$H_0 = \sum_i \left( \frac{m_i^2}{2b} + \frac{b\omega_0^2 r^2}{2} \right), \quad (5)$$

so that the part of the Hamiltonian associated with the interaction can be written as follows:

$$V = - \sum_{ik} J_{ik} m_i m_k + \sum_i \frac{u m_i^4}{8}.$$

In Eqs. (2) and (3) we shall assume subsequently that

$$H_0 = -\Delta \sum_i S_i^x, \quad V = - \sum_{ik} J_{ik} S_i^z S_k^z, \quad (6)$$

$$H_0 = -h \sum_i S_i^z, \quad V = - \sum_{ik} J_{ik} S_i S_k.$$

It is well known that the  $S$  matrix has the following form in the Keldysh technique:

$$S_c = T_c \exp \left\{ -i \int_c V(t) dt \right\}, \quad (7)$$

where  $T_c$  denotes ordering over a contour  $c$  shown in Fig. 1, which first goes from  $t = -\infty$  to  $t = +\infty$  and then in the opposite direction. This contour can be split into two,  $c_+$  and  $c_-$ , as shown in Fig. 1.

The Green function is defined as a matrix in terms of  $m(t_\pm)$ :

$$D_{\alpha\beta}(t-t') = -i \langle T_c \{ m(t_\alpha) m(t'_\beta) S_c \} \rangle; \quad \alpha, \beta = \pm. \quad (8)$$

The rules for the Keldysh diagram technique are the same as usual. The difference from the conventional technique at zero temperature (see, for example, Ref. 19) is an allowance for the matrix structure  $D_{\alpha\beta}(t-t')$ . In each order of perturbation theory we have, instead of the usual product of the Green functions, a sum of these functions and  $\alpha = +$  is assigned a symbol  $+1$ , whereas  $\alpha = -$  is assigned  $-1$ . For example, the graph in Fig. 2 has the following form (the usual factors are omitted):

$$\sum_{\xi} (\sigma_z)_{\xi\xi} \int D_{\xi\alpha}(t-t_1) D_{\xi\beta}(t-t_2) D_{\xi\gamma}(t-t_3) D_{\xi\delta}(t-t_4) dt, \quad (9)$$

$$(\sigma_z)_{\xi\xi} = \begin{cases} 1, & \xi = + \\ -1, & \xi = - \end{cases}.$$

where

$$S = T \exp \left\{ -i \int_{-\infty}^{\infty} dt \sum_{\alpha} (\sigma_z)_{\alpha\alpha} V_{\alpha}(t) \right\},$$

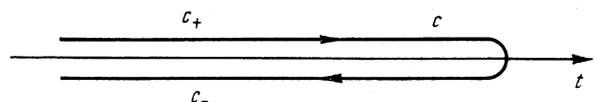


FIG. 1.

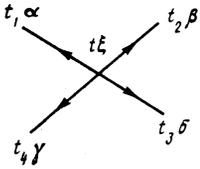


FIG. 2.

We can easily see that perturbation theory can be reformulated in terms of the usual  $S$  matrix. We shall assume that

$$V_\alpha(t) = V\{m_\alpha(t)\}, \quad (10)$$

$$m_\alpha(t) = m(t_\alpha).$$

It should be noted that the system (10) contains essentially two fields  $m_+(t)$  and instead of a complex profile  $c = c_+ + c_-$ , we obtain one simple profile  $c_+$ . Naturally, perturbation theory should also be developed for two fields  $m_\pm$ , but with one contour. This can be done quite easily, as follows. The zeroth approximation is a function  $D_{\alpha\beta}^{(0)}(t-t')$  which is obtained from Eq. (8) by substituting  $S_c = 1$ . We shall write down this function for the zeroth Hamiltonian of Eq. (5). However, we must note first that in the most general case we have<sup>14</sup>

$$D_{++} = \frac{1}{2}F + \frac{1}{2}(D^R + D^A), \quad D_{--} = \frac{1}{2}F - \frac{1}{2}(D^R + D^A),$$

$$D_{+-} = \frac{1}{2}F - \frac{1}{2}(D^R - D^A), \quad D_{-+} = \frac{1}{2}F + \frac{1}{2}(D^R - D^A), \quad (11)$$

$$F = -2iK,$$

where  $K$  is the correlation function of the field  $m(t)$ , whereas  $D^R$  and  $D^A$  are the retarded and advanced Green functions. In the  $\omega$  representation we find that the fluctuation-dissipation theorem is satisfied under equilibrium conditions:

$$\text{Im } D^R(\omega) = -\text{th } \frac{\omega}{2T} K(\omega). \quad (12)$$

It is clear from Eq. (5) that

$$K_0(\omega) = \frac{1}{2}\pi b\omega \text{cth } \frac{\omega}{2T} [\delta(\omega - \omega_0) + \delta(\omega + \omega_0)],$$

$$D_0^R(\omega) = \frac{1}{2}b\omega_0 \{1/(\omega - \omega_0 + i\delta) - 1/(\omega + \omega_0 + i\delta)\}, \quad (13)$$

$$D_0^A(\omega) = \{D_0^R(\omega)\}^*.$$

Once the zeroth Green functions are defined in terms of (11) and (13), the complete Green functions can be calculated from

$$D_{\alpha\beta}(t-t') = -i\langle T\{m_\alpha(t)m_\beta(t')S\} \rangle, \quad (14)$$

where  $T$  denotes the usual time ordering and  $S$  is defined by Eq. (10). Perturbation theory developed on the basis of Eqs. (7), (8), and (13) is fully identical with perturbation theory based on Eqs. (10), (13), and (14). However, the latter is more convenient.

We must now average the  $S$  matrix in Eq. (10), with allowance for Eq. (4), over the exchange integrals  $J_{ik}$ . We begin, however, by discussing the general procedure for averaging over random external fields that in general may depend on time.

We shall assume that our field  $m(t)$  interacts with a Gaussian random external field  $\varphi(t)$  characterized by the interaction Hamiltonian

$$V(\varphi, m) = g\varphi(t)m^2(t), \quad (15)$$

$$\langle \varphi(t)\varphi(t') \rangle = L(t-t').$$

The  $S$  matrix of Eq. (10) with  $V$  from Eq. (15) can in general be averaged over the random field  $\varphi(t)$ , because there is no denominator in the case of the  $S$  matrix of Eq. (10). Averaging Eq. (10) over  $\varphi(t)$ , we obviously obtain

$$S = T \exp \left\{ -\frac{g^2}{2} \iint dt dt' L(t-t') \right. \\ \left. \times \sum_{\alpha\beta} (\sigma_z)_{\alpha\alpha} (\sigma_z)_{\beta\beta} m_\alpha^2(t) m_\beta^2(t') \right\}. \quad (16)$$

We shall now show that any loop obtained from Eq. (16) vanishes exactly. This means that in reality all the loops corresponding to an external field, i.e., vacuum loops, vanish and the correlation function of the external field is not renormalized, as should be the case. The simplest loop can be expressed in terms of a time integral of an expression proportional to

$$\sum_{\alpha\beta} (\sigma_z)_{\alpha\alpha} (\sigma_z)_{\beta\beta} D_{\alpha\beta}^2(t-t'). \quad (17)$$

Using the explicit expression for  $D_{\alpha\beta}$  in Eq. (11) and the simple fact that

$$D^R(t)D^A(t) = 0, \quad (18)$$

we can easily show that Eq. (17) does indeed vanish. This happens because the external field  $\varphi(t)$  does not have the index  $\alpha$  and, consequently, the correlation function  $L(t)$  is independent of  $\alpha$  and  $\beta$ . A similar mechanism causes vanishing of any vacuum loop.

We must note a considerable difference between the interaction of Eq. (15) with a random external field and the analogous interaction with a Gaussian random field which is in thermodynamic equilibrium. In the latter case the  $S$  matrix can be averaged, in its general form, over the field  $\varphi$ , and instead of Eq. (16), we now obtain

$$S = T \exp \left\{ -\frac{ig^2}{2} \iint dt dt' \sum_{\alpha\beta} (\sigma_z)_{\alpha\alpha} (\sigma_z)_{\beta\beta} L_{\alpha\beta}(t-t') \right. \\ \left. \times m_\alpha(t) m_\beta(t') \right\}, \quad (19)$$

$$L_{\alpha\beta}(t-t') = -i\langle T[\varphi_\alpha(t)\varphi_\beta(t')] \rangle.$$

Equation (19) differs from Eq. (16) only because  $L_{\alpha\beta}$  is now strongly dependent on the indices  $\alpha$  and  $\beta$ . The components of  $L_{\alpha\beta}$  can be expressed in analogy with Eq. (11), in terms of the correlation function of  $L$  and  $G^R$ ,  $G^A$  for the field  $\varphi(t)$ . We obtain Eq. (16) if we ignore  $G^R$  and  $G^A$ . Therefore, a thermodynamic-equilibrium Gaussian field differs from an external random Gaussian field by the presence of  $G^R$  and  $G^A$ , which describe the reaction of the field  $\varphi(t)$  on the interaction of the field  $m(t)$  and give rise to renormalization of the correlation function  $L_{\alpha\beta}(t)$ . In the case of the usual random field  $\varphi(t)$  there is no such reaction in Eqs. (15) and (16) and, therefore, the field  $\varphi(t)$  is "insensitive" to  $m(t)$ .

We can thus see that averaging over external fields in the  $S$  matrix of Eq. (10) is a trivial matter. This applies also

to the averaging over static random quantities such as over the random exchange interaction  $J_{ik}$ . No replicas appear and instead we have a nonlocality on the time scale, which is similar to that which appeared in Eq. (16).

Substituting  $V$  from Eq. (5) into Eq. (10), and averaging over  $J_{ik}$ , we find from Eq. (4) that

$$S = T \exp \left\{ - \iint dt dt' \sum_{i h \alpha \beta} (\sigma_z)_{\alpha\alpha} (\sigma_z)_{\beta\beta} I_{ik} m_i^\alpha(t) \times m_k^\alpha(t) m_i^\beta(t') m_k^\beta(t') - i \int dt \sum_{i\alpha} (\sigma_z)_{\alpha\alpha} \frac{u(m_i^\alpha)^4}{8} \right\}. \quad (20)$$

As already pointed out in the Introduction, we shall consider only the molecular field theory. A standard decoupling corresponding to this approximation makes it possible to derive from Eq. (20) the molecular field equations for our problem, which are in fact equations for the correlation function  $D_{\alpha\beta}$ :

$$D_{\alpha\beta}(t-t') = -i \langle T \{ m_\alpha(t) m_\beta(t') S \} \rangle, \\ S = T \exp \left\{ -2iI_0 \iint dt dt' \sum_{\alpha\beta} (\sigma_z)_{\alpha\alpha} (\sigma_z)_{\beta\beta} D_{\alpha\beta}(t-t') \times m_\alpha(t) m_\beta(t') - i \int dt \sum_{\alpha} (\sigma_z)_{\alpha\alpha} \frac{u m_\alpha^4}{8} \right\}, \quad (21)$$

$$I_0 = \sum_h I_{ih}.$$

The averaging in the system (21) is carried out over the zeroth Hamiltonian  $H_0$  defined by Eq. (5).

We shall now show that the system of the molecular field equations (21) has a very simple physical meaning. We note that, according to Eq. (19) and the arguments discussed used to derive Eq. (19), the system (21) describes the case when our field  $m(t)$  interacts with a certain dynamic equilibrium quantum field  $h(t)$ . In this case the Hamiltonian for the field  $m$  and the  $S$  matrix are as follows:

$$H = -hm + U(m, r), \\ S = T \exp \left\{ -i \int dt \sum_{\alpha} (\sigma_z)_{\alpha\alpha} \left[ -h_\alpha(t) m_\alpha(t) + \frac{u}{8} m_\alpha^4(t) \right] \right\}, \\ L_{\alpha\beta}(t-t') = -i \langle T (h_\alpha(t) h_\beta(t')) \rangle = 4I_0 D_{\alpha\beta}(t-t'). \quad (22)$$

The last line of Eq. (22) contains a correlation of an equilibrium field  $h$ , similar to  $L_{\alpha\beta}$  in Eq. (19), and this equation shows how the former is related to the field correlation function  $D_{\alpha\beta}(t)$ .

We shall now introduce prior averaging over the configurations  $J_{ik}$  a quantum molecular field:

$$h_i(t) = 2 \sum_k J_{ik} m_k(t). \quad (23)$$

It is clear from Eq. (23) that in the zeroth molecular-field approximation, i.e., in the case of a large number of the nearest neighbors and when Eq. (4) is obeyed, the field  $h(t)$  is indeed a Gaussian thermodynamic quantity with the correlation function  $L_{\alpha\beta}(t)$  of Eq. (22).

It therefore follows that our molecular field equations describe a very simple situation: the system contains random molecular fields  $h_i(t)$  which fluctuate in time. Such a picture differs radically from the usual situation, such as that encountered in a ferromagnet when molecular fields do not fluctuate in time but are essentially static quantities.

### 3. MOLECULAR FIELD EQUATIONS UNDER NONERGODIC CONDITIONS

The principal molecular field equations, i.e., the system (21), are valid in the ergodic and nonergodic regions. However, in the nonergodic region we must make an additional approximation, namely we have to assume that our system has a continuous spectrum of exponentially long relaxation times. This assumption has been discussed in detail earlier<sup>1,3,9-13,16,17</sup> for steady-state dynamics of classical spin glasses. Since nonergodic times are related to intervalley transitions and these, as pointed out by the present author in Ref. 4, depend weakly on whether the conditions are of the quantum kind, it is clear that the assumption of a continuous spectrum of nonergodic relaxation times should be made also in the quantum case. Following Refs. 1, 3, and 13, we shall assume that

$$D_{\alpha\beta}(t) = D_{\alpha\beta}^{(0)}(t) + D_{\alpha\beta}^{(s)}(t), \\ F(t) = F_0(t) - 2iq(z), \\ D^R(t) = D_0^R(t) - \frac{\alpha}{|t|T} \Delta'(z) \vartheta(t), \\ D^A(t) = D_0^A(t) - \frac{\alpha}{|t|T} \Delta'(z) \vartheta(-t), \\ z = \alpha \ln \frac{|t|}{\tau}, \quad \alpha \rightarrow 0, \quad z \sim 1. \quad (24)$$

In the system (24), we separate from  $D_{\alpha\beta}$  the regular part  $D_{\alpha\beta}^{(0)}(t)$  from the singular part  $D_{\alpha\beta}^{(s)}(t)$ . Consequently,  $F$ ,  $D^R$ , and  $D^A$  split into regular parts  $F_0$ ,  $D_0^R$ , and  $D_0^A$ , and singular parts related to  $q(z)$  and  $\Delta(z)$ , which are functions of a logarithmic variable  $z$ . These are unknown functions of the theory and they can be described by equations which are fully equivalent to the corresponding equations in Refs. 3 and 13 [see Eq. (46)]. In the system (24) the last line defines the conditions for going to the limit in the case of the principal logarithms,  $\vartheta(t) = 0$  when  $t < 0$  and  $\vartheta(t) = 1$  when  $t > 0$ , where  $\tau$  is the paramagnetic time and  $T$  is the temperature.

The quantity  $D_{\alpha\beta}^{(0)}(t)$  in the system (24) changes after a characteristic microscopic time, whereas  $D_{\alpha\beta}^{(s)}(t)$  changes after a macroscopic time

$$\tau_0 = \tau \exp(1/\alpha). \quad (25)$$

If  $\alpha \rightarrow 0$ , the times  $\tau$  and  $\tau_0$  are parametrically separate. However, the nonergodicity is not manifested by slow relaxation after a macroscopic time  $\tau_0$ , but by the breakdown of the fluctuation-dissipation theorem in the case  $D_{\alpha\beta}^{(s)}(t)$ . It is the failure of this theorem that is a criterion of nonergodicity (for details see Refs. 1, 3, and 9-13). It should also be noted that the variable  $z$  satisfies the ultrametry condition

$$z(t_1 - t_3) = \max\{z(t_1 - t_2), z(t_2 - t_3)\} \quad (26)$$

for any macroscopic time  $t_1$ ,  $t_2$ , or  $t_3$ . We can allow for the

ultrametry property of the variable  $z$  by a procedure similar to that used in Refs. 1, 3, and 13. Let us consider the following time interval:

$$[t_0, t_1], \quad t_0 \rightarrow -\infty, \quad t_1 \rightarrow +\infty. \quad (27)$$

We shall separate this interval into  $j^{k+1}$  subintervals of duration  $\tau$ :

$$t_1 - t_0 = \tau j^{k+1}. \quad (28)$$

The size of the subinterval  $\tau$  is selected to be much greater than any microscopic time ( $\omega_0 \tau \gg 1$ ,  $\Delta \tau \gg 1$ ,  $\gamma \tau \gg 1$ ), where  $\omega_0$  and  $\Delta$  are the parameters of the Hamiltonians (1) and (2), whereas  $\gamma$  is any damping which may occur during microscopic time intervals. Any instant during the interval (27) can be represented as follows:

$$\begin{aligned} t &= t_a + \tilde{t}; \quad \tilde{t} \in [-\tau/2, \tau/2], \\ t_a &= t_0 + \tau(a_0 j^k + a_1 j^{k-1} + \dots + a_{j-1} j + a_j), \\ a_l &= 0, \dots, j-1; \quad l=0, \dots, k, \end{aligned} \quad (29)$$

where  $\tilde{t}$  changes in the interval  $[-\tau/2, \tau/2]$ . It follows from the definition of  $z$  in Eq. (24) that  $D_{\alpha\beta}^{(s)}(t)$  changes after a time much longer than  $\tau$ , while  $D_{\alpha\beta}^{(0)}(t)$  still changes after a time much shorter than  $\tau$ . Therefore, we have  $D_{\alpha\beta}^{(0)}(t) = 0$ , if  $t_a \neq t_b$  for any  $t$  and  $t'$  and differs from zero only for  $a = b$ . On the other hand,  $D_{\alpha\beta}^{(s)}(t)$  does not change during such time intervals. If we label instants of the time by  $a$  and  $\tilde{t}$ , we obtain

$$D_{\alpha\beta}^{(s)}(t-t') = D_{\alpha\beta ab}^{(s)}, \quad (30)$$

$$D_{\alpha\beta}^{(0)}(t-t') = D_{\alpha\beta}^{(0)}(\tilde{t}-\tilde{t}') \delta_{ab}.$$

Such a representation is fully analogous to the corresponding representation in the static theory of Ref. 4, but the meaning of the index  $a$  is now naturally different. We can see from Eq. (29) that  $t_a$  is governed by  $k+1$  numbers  $a_j$ . Let us assume that we now have  $j \rightarrow \infty$ , so that since  $a_l \sim j$ , we have

$$\begin{aligned} z(t_a - t_b) &= z_l = (k-l+1)\Delta z, \\ z(t_1 - t_0) &= z_{\max} = z_0 = (k+1)\Delta z, \end{aligned} \quad (31)$$

$$\begin{aligned} \Delta z &= \alpha \ln j \rightarrow 0, \quad k \rightarrow \infty, \quad j \rightarrow \infty, \\ \alpha &\rightarrow 0, \quad \Delta z/\alpha \rightarrow \infty, \quad z_{\max} \rightarrow \infty. \end{aligned}$$

It is clear from the system (31) that  $z(t_a - t_b)$  considered in the limit  $j \rightarrow \infty$  is governed only by the number  $l$  in the hierarchical ladder, but not by the specific values of  $a_j$ . This implies ultrametry.

We shall now substitute Eqs. (24) and (30) into Eq. (21). Then, in the exponential function in Eq. (21) we find two terms:

$$\begin{aligned} I_1 &= \sum_{\alpha\beta} q_{ab} \left( \int_{-\tau/2}^{\tau/2} d\tilde{t} m_{\alpha\alpha}(\tilde{t}) \right) \left( \int_{-\tau/2}^{\tau/2} d\tilde{t} m_{\beta\beta}(\tilde{t}) \right) \\ &+ \sum_{\alpha\beta} p_{ab} \left( \int_{-\tau/2}^{\tau/2} d\tilde{t} m_{\alpha\alpha}(\tilde{t}) \right) \left( \int_{-\tau/2}^{\tau/2} d\tilde{t} m_{\alpha\beta}(\tilde{t}) \right), \\ m_{\alpha\alpha}(\tilde{t}) &= m_{\alpha}(t_a + \tilde{t}), \\ q_{ab} &= q[z(t_a - t_b)], \end{aligned} \quad (32)$$

$$p_{ab} = \frac{\alpha}{2T|t_a - t_b|} \Delta' [z(t_a - t_b)] (1 - \delta_{ab}),$$

$$I_2 = \sum_{\alpha\beta} \int_{-\tau/2}^{\tau/2} d\tilde{t}_1 \int_{-\tau/2}^{\tau/2} d\tilde{t}_2 D_{\alpha\beta}^{(0)}(\tilde{t}_1 - \tilde{t}_2) m_{\alpha\alpha}(\tilde{t}_1) m_{\beta\beta}(\tilde{t}_2).$$

The first term  $I_1$  stems from the singular part  $D_{\alpha\beta}^{(s)}$ , and the second from the regular part. The term with  $D_{\alpha\beta}^{(s)} - D_{\alpha\beta}^{(0)}$  will be ignored because it is small in the zeroth order when expressed in terms of the principal logarithms. It should be noted that averaging using the  $S$  matrix from Eq. (32) should be carried out over the zeroth Hamiltonian of Eqs. (5) and (6). Then, the correlation functions decrease after time intervals of the order of  $\omega_0^{-1}$  and  $\Delta^{-1}$ . We can therefore assume that  $m_{\alpha\alpha}(\tilde{t})$  and  $m_{\alpha\beta}(\tilde{t})$  in Eq. (32) commute. For the same reasons the integrals with respect to  $\tilde{t}$  can be extended over an interval  $[-\infty, \infty]$ . We finally obtain from Eq. (21) ( $\tilde{t}, \tilde{t}' \rightarrow t, t'$ ):

$$D_{\alpha\beta}^{(0)}(t-t') = -i \langle T \{ m_{\alpha\alpha}(t) m_{\beta\beta}(t') S \} \rangle,$$

$$D_{\alpha\beta\sigma\delta}^{(s)} = -i \langle m_{\alpha\alpha}(t) m_{\beta\beta}(t') S \rangle_{\alpha \neq \beta}, \quad (33)$$

$$\begin{aligned} S &= T \exp \left\{ -2iI_0 \iint dt dt' \sum_{\alpha\beta} (\sigma_z)_{\alpha\alpha} (\sigma_z)_{\beta\beta} \sum_{ab} [D_{\alpha\beta ab}^{(s)} \right. \\ &+ \delta_{ab} D_{\alpha\beta}^{(0)}(t-t')] m_{\alpha\alpha}(t) m_{\beta\beta}(t') - i \int dt \sum_{\alpha} (\sigma_z)_{\alpha\alpha} \frac{um_{\alpha\alpha}^4}{8} \left. \right\}. \end{aligned}$$

In Eq. (33) the  $T$  product denotes ordering in respect of  $t$ , but not in respect of  $a$ , because  $m_a(t)$  and  $m_b(t)$  commute. For the same reason the expression for  $D^{(s)}$  is simplified by dropping the  $T$ -product symbol, so that  $D^{(s)}$  is independent in fact of  $t$  and  $t'$ , whereas in the term  $D_{\alpha\beta ab}^{(s)}$  the  $T$ -ordering symbol in the  $S$  matrix orders the operators and  $m_{\alpha\beta}(t')$  separately in respect of  $t$  and  $t'$ . However, the average in  $D_{\alpha\beta ab}^{(s)}$  does not then vanish, because the  $S$  matrix contains terms which mix  $m_{\alpha\alpha}$  and  $m_{\beta\beta}$ .

The system (33) is a complete set of equations for  $D_{\alpha\beta}^{(0)}(t)$  and  $D_{\alpha\beta}^{(s)}$ . It should be noted that essentially the system (33) represents a system of equations for a set of commuting fields  $m_a(t)$ . Such separation into commuting fields  $m_a(t)$  is analogous to that adopted in Ref. 4, but in the present case it is important that the index  $a$  denotes a macroscopic instant of time  $t_a$ . In the case of such commuting fields the zeroth Hamiltonian is

$$H_0 = \sum_{\alpha} H_0(m_{\alpha}, r_{\alpha}), \quad (34)$$

where  $H_0(m, r)$  is defined in Eq. (5). It should be noted that, in spite of the fact that  $m_a(t)$  commute, the indices  $\alpha$  and  $\beta$  are retained and they will be very important later.

Standard Stratonovich-Hubbard transformations<sup>1,4,13</sup> yield the following expression for the  $S$  matrix:

$$\begin{aligned} S &= \left\langle T \exp \left\{ i \sum_{\alpha_0 \dots \alpha_k} (\sigma_z)_{\alpha\alpha} \xi_{\alpha_0 \dots \alpha_k}^{(k)} \int dt m_{\alpha_0 \dots \alpha_k}(t) \right. \right. \\ &\left. \left. - 2iI_0 \iint dt dt' \sum_{\alpha\beta} (\sigma_z)_{\alpha\alpha} (\sigma_z)_{\beta\beta} D_{\alpha\beta}^{(0)}(t-t') m_{\alpha\alpha}(t) \right. \right. \end{aligned}$$

$$\times m_{\beta\alpha}(t') - i \int dt \sum_{\alpha\alpha} (\sigma_z)_{\alpha\alpha} \frac{um_{\alpha\alpha}}{8} \Bigg\}_{h, \xi},$$

$$\xi_{\alpha_0 \dots \alpha_{k-1}}^{(k)} = h^{(0)} + h_{\alpha_0}^{(1)} + \dots + h_{\alpha_0 \dots \alpha_{k-1}}^{(k)} + \xi_{\alpha_0}^{(0)} + \xi_{\alpha_0 \alpha_1}^{(1)} + \dots + \xi_{\alpha_0 \dots \alpha_{k-1}}^{(k)}.$$

(35)

The symbol  $\langle \dots \rangle_{h, \xi}$  denotes averaging over all the variables  $h$  and  $\xi$  which occur in Eq. (35). This averaging can be carried out explicitly as follows (see, for example, the analogous expression in Ref. 4):

$$\langle \dots \rangle_{h, \xi} = \int \frac{dh^{(0)}}{(8\pi I_0 q_0)^{1/2}} \exp \left\{ -\frac{(h^{(0)})^2}{8I_0 q_0} \right\}$$

$$\dots$$

$$\times \prod_{\alpha_0} \int \frac{dh_{\alpha_0}^{(1)}}{(8\pi I_0 q_1)^{1/2}} \exp \left\{ -\frac{(h_{\alpha_0}^{(1)})^2}{8I_0 q_1} \right\}$$

$$\dots$$

$$\times \prod_{\alpha_0 \dots \alpha_{k-1}} \int \frac{dh_{\alpha_0 \dots \alpha_{k-1}}^{(k)}}{(8\pi I_0 q_k)^{1/2}} \exp \left\{ -\frac{(h_{\alpha_0 \dots \alpha_{k-1}}^{(k)})^2}{8I_0 q_k} \right\}$$

$$\times \int \frac{d\xi_{\alpha_0}^{(0)} d\xi_{\alpha_0 \alpha_1}^{(1)}}{(8\pi/\tau T) I_0 (-\Delta_0'/p_0)} \exp \left\{ -i p_0 \tau T \frac{(\xi_{\alpha_0}^{(0)})^2 - (\xi_{\alpha_0 \alpha_1}^{(1)})^2}{8I_0 (-\Delta_0')} \right\}$$

$$\dots$$

$$\times \prod_{\alpha_0 \dots \alpha_{k-1}} \int \frac{d\xi_{\alpha_0 \dots \alpha_{k-1}}^{(k)}}{(8\pi/\tau T) I_0 (-\Delta_k'/p_k)}$$

$$\times \exp \left\{ -i p_k \tau T \frac{(\xi_{\alpha_0 \dots \alpha_{k-1}}^{(k)})^2 - (\xi_{\alpha_0 \dots \alpha_{k-1}}^{(k)})^2}{8I_0 (-\Delta_k')} \right\} (\dots). \quad (36)$$

Here,

$$q_i' = q_i - q_{i-1}, \quad \Delta_i' = \Delta_{i+1} - \Delta_i,$$

$$q_i = q(z_i), \quad \Delta_i = \Delta(z_i), \quad p_i = j^{k-1+i},$$

and the dots in Eq. (36) denote products of intermediate numbers of variable expressions similar to those written down explicitly.

Equations (35) and (36) are very similar to the corresponding expressions in Ref. 2, but there are two very important differences. Firstly, instead of fictitious replicas, we are dealing here with real indices  $\alpha, \beta = +, -$ , corresponding to two different time profiles in the density matrix. Secondly, as before—and in contrast to Ref. 4—we are dealing with an  $S$  matrix and we can therefore consider processes far from equilibrium.

#### 4. INTRODUCTION OF THE FREE ENERGY AND THE EXPRESSION FOR THE $S$ MATRIX IN TERMS OF A COMPLETE PROBABILITY FUNCTIONAL

It is known from Refs. 1, 2, 4, and 13 that the basis of the theory of nonergodicity is provided by partial differential equations for the free energy, magnetization, and various distribution functions. In particular, we can calculate a complete probability functional of molecular fields.<sup>1,13</sup> Using the results of Refs. 1, 4, and 13 we can write down directly the necessary expressions. However, we must bear in mind the following. Our technique generally differs from that used in Refs. 1, 4, and 13. However, in spite of this technical difference, all the recurrence relationships, partial differential equations, and distribution functions for the static case are

identical with the results of Ref. 4. We shall not give here the fairly cumbersome derivations demonstrating this point, but consider simply one very important point, namely the determination of the free energy and of other thermodynamic quantities in the Keldysh technique. We shall do this by considering a specific example. Let us assume that  $H_0(m, r)$  is obtained from Eq. (5) and the interaction is described by

$$V = -hm + um^4/8, \quad (37)$$

where the  $S$  matrix is found using Eqs. (10) and (37). We shall now calculate the magnetization  $M(h)$  for the complete Hamiltonian. It is quite obvious that

$$M(h) = \langle m_{\alpha}(t) S \rangle \quad (38)$$

and that it is independent of  $\alpha$  and  $t$ . On the other hand, if the complete range of integration in terms of  $t$  is given by Eq. (27), it is obvious that

$$\frac{\partial}{\partial h} \langle S(h) \rangle = i(t_1 - t_0) M(h) \sum_{\alpha} (\sigma_z)_{\alpha\alpha}. \quad (39)$$

In Eq. (39) the left- and right-hand sides are expressions proportional to

$$\sum_{\alpha} (\sigma_z)_{\alpha\alpha} = 0,$$

whereas  $M(h)$  is finite. It follows from Eq. (39) that

$$\langle S(h) \rangle = 1 + i(t_1 - t_0) \varphi(h) \sum_{\alpha} (\sigma_z)_{\alpha\alpha},$$

$$M(h) = \partial \varphi(h) / \partial h. \quad (40)$$

Equation (40) gives the free energy  $\varphi(h)$  as a coefficient in front of an expression which vanishes. Similarly, in the replica method<sup>1,2</sup> the free energy  $\varphi(h)$  is defined as the coefficient in front of  $n$  ( $n$  is the number of replicas and we assume that  $n \rightarrow 0$ ). It should be noted that  $\varphi(h)$  is the difference between the total free energy and that free energy which would be obtained in the absence of the interaction, but in the derivation of the recurrence relationships this is not important because the latter energy is independent of  $h$ .

Using the definition of the free energy given by Eq. (40), we can now deduce the usual recurrence relationships. In the intermediate stages we have to allow for the property

$$\sum_{\alpha} (\sigma_z)_{\alpha\alpha} = 0, \quad (41)$$

by analogy with the procedure used to derive the recurrence relationships in Refs. 1 and 13, where it is assumed that  $n \rightarrow 0$ . In general, Eq. (41) plays exactly the same role in quantum dynamics as the property  $n \rightarrow 0$  in the conventional theory. This is why we can avoid the use of replicas. However, it should be pointed out that the property described by Eq. (41) is inherently a feature of the Keldysh technique applied to any system, whereas the replica methods can be used only in the case of disordered systems.

Fairly cumbersome calculations, fully analogous to those described in Refs. 1 and 13, allows us to calculate—for example—the moment  $M(h)$ . It is expressed in terms of the limiting value of the function  $M(z, h)$  ( $M(h) = M(\infty, h)$ ) that satisfies the following simple differential equation:

$$-\frac{\partial M(z, h)}{\partial z} = 4I_0 \left\{ \frac{1}{2} q'(z) \frac{\partial^2 M(z, h)}{\partial h^2} - \frac{1}{T} \Delta'(z) M(z, h) \frac{\partial M(z, h)}{\partial h} \right\},$$

$$M(0, h) = M_0(h) = \langle m_{\alpha\alpha} S_{0\alpha}(h) \rangle,$$

$$S_{0\alpha}(h) = T \exp \left\{ ih \sum_{\alpha} (\sigma_z)_{\alpha\alpha} \int dt m_{\alpha\alpha}(t) - 2iI_0 \sum_{\alpha\beta} (\sigma_z)_{\alpha\alpha} \times (\sigma_z)_{\beta\beta} \iint dt dt' D_{\alpha\beta}^{(0)}(t-t') m_{\alpha\alpha}(t) m_{\beta\beta}(t') - i \sum_{\alpha} (\sigma_z)_{\alpha\alpha} \int dt \frac{um_{\alpha\alpha}^4(t)}{8} \right\}. \quad (42)$$

We can show that  $M(h)$  in Eq. (42) is identical with the corresponding expression in Ref. 4.

We can also express the complete  $S$  matrix in Eq. (35) in terms of the complete probability functional  $F(h_a)$ :

$$S = \int Dh_a F(h_a) \prod_{\alpha} S_{0\alpha}(h_a), \quad (43)$$

$$Dh_a = \prod_{\alpha} dh_a.$$

The complete probability functional  $F(h_a)$  is a function of all the fields considered at macroscopic times  $h_a$  and has the following form<sup>1,13</sup>:

$$F(h_a) = \int dh^{(0)} \frac{1}{(8\pi I_0 q_0)^{1/2}} \exp \left\{ -\frac{(h^{(0)} - h)^2}{8I_0 q_0} \right. \\ \times \left\{ \prod_{\alpha_0} \int dh_{\alpha_0}^{(1)} P(z_0, h_{\alpha_0}, z_1, h_{\alpha_0}^{(1)}) \right. \\ \times \prod_{\alpha_0, \alpha_1} \int dh_{\alpha_0, \alpha_1}^{(2)} P(z_1, h_{\alpha_0}^{(1)}, z_2, h_{\alpha_0, \alpha_1}^{(2)}) \\ \dots \\ \times \prod_{\alpha_0, \dots, \alpha_{k-1}} \int dh_{\alpha_0, \dots, \alpha_{k-1}}^{(k)} P(z_{k-1}, h_{\alpha_0, \dots, \alpha_{k-2}}, z_k, h_{\alpha_0, \dots, \alpha_{k-1}}^{(k)}) \\ \times \prod_{\alpha_0, \dots, \alpha_k} \delta(h_{\alpha_0, \dots, \alpha_k} - h_{\alpha_0, \dots, \alpha_{k-1}}^{(k)}) \left. \right\} \\ z_0 = z_{\max} = k\Delta z; \quad z_k = z_{\min} = \Delta z, \quad (44)$$

where  $h$  is an external magnetic field and  $P(z, h, z', h')$  is the transient probability of some Markov process and satisfies the Kolmogorov equations in terms of the first and second variables<sup>1,13</sup>:

$$-\frac{\partial P}{\partial z} = 4I_0 \left\{ \frac{1}{2} q'(z) \frac{\partial^2 P}{\partial h^2} - \frac{1}{T} \Delta'(z) M(z, h) \frac{\partial P}{\partial h} \right\}, \\ \frac{\partial P}{\partial z'} = 4I_0 \left\{ \frac{1}{2} q'(z') \frac{\partial^2 P}{\partial h'^2} + \frac{1}{T} \Delta'(z') \frac{\partial}{\partial h'} [PM(z', h')] \right\}, \\ P(z, h, z, h') = \delta(h - h'). \quad (45)$$

The system of equations (42)–(45) solves in principle the problem of steady-state dynamics in the quantum case. It

has to be supplemented also by an equation for  $D_{\alpha\beta}^{(0)}(t)$ , which will be given below.

Equation (43) can be used to calculate any correlation function. We recall that  $F(h_a)$  and  $P(z, h, z', h')$  are normalized to unity and also that  $\langle S_{0\alpha}(h_a) \rangle = 1$ . We can therefore write down, for example, a correlation function

$$q(z_{ab}) = \int Dh_a F(h_a) \prod_{\alpha} \langle m_{\alpha\alpha} m_{\beta\beta} S_{0\alpha}(h_a) \rangle \\ = \int dh_1 dh_2 F_2(z_{ab}, h_1, h_2) M_0(h_1) M_0(h_2), \quad (46)$$

where  $F_2(z_{ab}, h_1, h_2)$  is defined in terms of an integral of  $F(h_a)$  with respect to all values of  $h_c$ , except  $h_a$  and  $h_b$ . Exactly similarly we have

$$D_{\alpha\beta}^{(0)}(t-t_1) = \int dh F_1(h) D_{\alpha\beta}^{(0)}(h, t-t_1), \quad (47)$$

$$D_{\alpha\beta}^{(0)}(h, t-t_1) = -i \langle T m_{\alpha}(t) m_{\beta}(t_1) S_0(h) \rangle.$$

The system (47) closes our system of equations. Exactly the same procedure can be used to obtain any correlation function, for example, a four-particle function in which the times  $t_1$  and  $t_2$  lie within the same macroscopic interval  $[t_a - \tau/2, t_a + \tau/2]$ , whereas  $t_3$  and  $t_4$  lie in a different macroscopic interval  $[t_b - \tau/2, t_b + \tau/2]$ . Obviously, this correlation function depends on  $t_1 - t_2$ ,  $t_3 - t_4$ , and  $z_{ab}$ .

## 5. NONEQUILIBRIUM DISTRIBUTION FUNCTION OF MOLECULAR FIELDS

We can at long last tackle the main task, which is calculation of an arbitrary nonlinear susceptibility. We shall consider only the susceptibility at macroscopic times, which corresponds to slow (long-term) relaxation in the experiments. We shall consider only one example of a generalized nonlinear susceptibility, but its derivation will demonstrate the general method for calculation of such quantities.

In principle we could write down equations of type (21) under transient conditions, when the correlation functions  $D_{\alpha\beta}(t, t')$  no longer depend on the difference between the times, and we could solve these equations in the nonergodic range. This is a very difficult task and we have been unable to solve it in its general form. We have achieved something less, namely, we have assumed that  $q(z)$  and  $\Delta(z)$  are determined by a steady-state process and have calculated all possible nonequilibrium susceptibilities. Since the system exhibits slow relaxation and  $q(z)$  and  $\Delta(z)$  are modified in a time  $\tau_0$  defined in Eq. (25) even if  $z \ll 0$ , it is obvious that such an approach is valid in the case of short macroscopic times, as long as

$$[q(z) - q(0)]/q(0) \ll 1, \quad (48)$$

i.e., if the expression obtained above for the susceptibility corresponds to the zeroth approximation in terms of the parameter (48).

Let us assume that we are dealing with a steady-state process and that some macroscopic moment in time  $t_a$  a magnetic field of arbitrary magnitude  $h$  is applied. We shall be interested in various quantities, at a different macroscopic time  $t_b$ , for example, the magnetic moment or the correlation function at  $t_1$  and  $t_2$  within the interval  $[t_b - \tau/2, t_b + \tau/2]$ . In attempts to calculate directly these quantities,

for example the value of the usual linear susceptibility, we meet the same difficulty as in the classical theory of Refs. 1 and 13. We have to calculate averages of the  $\langle m_{\alpha\alpha}, m_{\beta\beta} S \rangle$  type and identify the dependence of such an average on  $\alpha$  and  $\beta$ . However, it is clear from Eq. (46) that—as in Refs. 1 and 13, these averages are independent of  $\alpha$  and  $\beta$  in the replica method. The dependence on  $\alpha$  and  $\beta$  appears in the next order in respect of a large parameter used in the steepest-descent method. A direct calculation of the corrections to this method is very difficult. Therefore, the problem was bypassed in Refs. 1 and 13 and we shall do this here. We shall multiply Eq. (35) by  $m_{\beta\beta}$  and differentiate the resultant expression with respect to  $h_{a_0 \dots a_{l-1}}^{(l)}$ , where  $l$  is a level in the hierarchy corresponding to  $z(t_a - t_b) = z_l$ . If we multiply the whole expression by an external field  $h$ , we obtain a correction to  $\langle m_{\beta\beta} S \rangle$ , necessitated by application of a magnetic field  $h$  at a moment  $t_a$ , i.e., we obtain the susceptibility multiplied by  $h$ . This susceptibility is

$$h \sum_{\alpha c} \langle m_{\alpha c} m_{\beta\beta} S \rangle (\sigma_z)_{\alpha\alpha}, \quad (49a)$$

where the sum over  $c$  extends to all values of  $c$  such that

$$z(t_c - t_b) < z_l = z(t_a - t_b). \quad (49b)$$

Following Refs. 1 and 13, we find that this correction is

$$h \int F(z_l, h_1) M'(z_l, h_1) dh_1. \quad (50)$$

It then readily follows from Refs. 1 and 13 that

$$M(z, h) = \int P(z, h, 0, h_1) M_0(h_1) dh_1, \quad (51)$$

where  $M_0(h)$  is defined by Eq. (42). It is clear from Eq. (42) that  $M_0(h) = \langle m_{\alpha\beta} S_{0\beta}(h) \rangle$  and, therefore, Eqs. (50) and (51) yield a correction of the first order in  $h$  to the distribution function of the molecular fields  $\Delta F_1(h_1)$ , which appears because an external magnetic field is applied at a moment  $t_a$ :

$$\Delta F_1(h_1) = h \int F(z, h_2) \frac{\partial}{\partial h_2} P(z, h_2, 0, h_1). \quad (52)$$

Naturally this correction depends only on  $z = z_l = z(t_a - t_b)$ .

We can easily calculate these corrections also in higher orders in  $h$ . We readily find that they are obtained if Eq. (35) is multiplied by  $m_{\beta\beta}$  and differentiated several times with respect to  $h_{a_0 \dots a_{l-1}}^{(l)}$ . If we use a simple expression<sup>1,13</sup>

$$F_1(h_1) = \int F(z, h_2) P(z, h_2, 0, h_1) dh_2, \quad (53)$$

we find that the complete nonequilibrium expression  $F_1^{\text{neq}}(z, h_1)$  is

$$F_1^{\text{(neg)}}(z, h_1) = \int dh_2 F(z, h_2) \sum_{n=0}^{\infty} \frac{h^n}{n!} \frac{\partial^n}{\partial h_2^n} P(z, h_2, 0, h_1) = \int dh_2 F(z, h_2) P(z, h_2 + h, 0, h_1). \quad (54)$$

Equation (54) is the explicit expression for the complete nonequilibrium single-particle distribution function in our simple case of an arbitrary constant magnetic field. The derivation of this expression demonstrates quite clearly how we can calculate many-particle distribution functions or a sin-

gle-particle distribution function when a magnetic field varies several times, and so on. This can be done quite simply, but the procedures are fairly cumbersome so that we shall not do that here.

We must mention one very important circumstance. It is clear from Eq. (54) that if the inequality of Eq. (48) is satisfied, a nonequilibrium distribution<sup>2</sup> function can be calculated explicitly if we know the steady-state solution of the problem. This is also true of other quantities.

## 6. SLOW RELAXATION OF A SPECTRAL PROFILE AND OF A TRIPLE CORRELATION FUNCTION

In this concluding section we shall give two examples of slow relaxation of spectral functions, which describe in particular also the spectral profile. One such task was discussed in an earlier paper.<sup>4</sup> We shall assume that we are dealing with a structural order-disorder glass. As shown in Ref. 4, the optical spectrum of this glass includes frequencies of the order of characteristic molecular fields  $I_0^{1/2}$ . This spectrum begins to relax on application of an external magnetic field. The results of the preceding section show quite clearly how to describe this relaxation. The retarded correlation function is the same as in Ref. 4:

$$D^R(z, h, \omega) = \int dh_1 F_1^{\text{(neq)}}(z, h_1) \times \frac{\langle R(h_1 + y, \omega) \rangle_y}{\langle 2 \operatorname{ch} \{ ((y + h_1)^2 + \Delta^2)^{1/2} / 2T \} \rangle_y} - \frac{2\pi q}{T} \delta(\omega),$$

$$R(h, \omega) = \frac{\operatorname{ch} \{ (h^2 + \Delta^2)^{1/2} / 2T \}}{2(h^2 + \Delta^2)} \left\{ \frac{h^2}{T} R_0(h, \omega) - 2\Delta^2 \frac{(h^2 + \Delta^2)^{1/2}}{\omega^2 - (h^2 + \Delta^2)} \operatorname{th} \frac{(h^2 + \Delta^2)^{1/2}}{2T} \right\},$$

$$q = q(z=0) = \int F_1^{\text{(neq)}}(z, h_1) M_0^2(h_1) dh_1, \quad (55)$$

$$\int \frac{d\omega}{2\pi} R_0(h, \omega) = 1,$$

where  $F_1^{\text{neq}}(z, h_1)$  is defined by Eq. (54) and the averaging over  $y$  represents averaging over equilibrium molecular fields obtained in the static approximation of Ref. 4. At low values of  $\Delta$  a good approximation is provided by

$$\langle \dots \rangle_y = \int \frac{dy}{(8\pi I_0 dT)^{1/2}} (\dots) \exp\left(-\frac{y^2}{8I_0 dT}\right), \quad (56)$$

where  $dT \approx \frac{1}{4}$ . In Eq. (55) the function  $R_0(h, \omega)$  is that part of  $R(h, \omega)$  which is governed by low frequencies  $\omega \lesssim \Delta$  and is normalized to unity. In particular, in  $R_0(h, \omega)$  we have a part corresponding to complete freezing at microscopic frequencies. This part cancels out the corresponding term  $\sim 2\pi q \delta(\omega)$  in Eq. (55). The correlation function is related to  $D^R$  by the fluctuation-dissipation theorem. Equation (55) is derived using the Hamiltonian (2) of a structural order-disorder glass.

We shall conclude by considering one other very interesting example relating to quantum properties of a Heisenberg spin glass. It is well known that in the purely static case the odd correlation functions of spins vanish for a paramagnet. However, in the dynamic case such correlation functions are finite because the equation of motion for spin in-

cludes a triple dynamic vertex. However, the existence of odd dynamic correlation functions gives rise to a number of very interesting phenomena, for example, those exhibited by the scattering of polarized neutrons in a paramagnet subjected to a magnetic field (see, for example, Refs. 20 and 21). One can expect a Heisenberg spin glass also to have a non-zero three-particle correlation function. We shall consider this question separately and give here only the explicit form of the retarded three-particle Green function. For simplicity, we shall consider only the static case. Relaxation phenomena will be discussed separately. As in the derivation of Eq. (55), we shall employ the static approximation. Using the Hamiltonian of Eq. (3), we find after fairly lengthy calculations

$$K_{\mu\nu\lambda}^R(\omega_1, \omega_2, \omega_3) = -\frac{i}{6} \varepsilon_{\mu\nu\lambda} \int d\mathbf{h} F_1(\mathbf{h}) \langle z(\mathbf{y}+\mathbf{h}) \rangle_{\mathbf{y}}^{-1} \times \langle R_{-+z}(\mathbf{y}+\mathbf{h}, \omega_1, \omega_2, \omega_3) - R_{-+z}(\mathbf{y}+\mathbf{h}, \omega_2, \omega_1, \omega_3) + R_{-+z}(\mathbf{y}+\mathbf{h}, \omega_3, \omega_1, \omega_2) - R_{-+z}(\mathbf{y}+\mathbf{h}, \omega_1, \omega_3, \omega_2) + R_{-+z}(\mathbf{y}+\mathbf{h}, \omega_2, \omega_3, \omega_1) - R_{-+z}(\mathbf{y}+\mathbf{h}, \omega_3, \omega_2, \omega_1) \rangle_{\mathbf{y}}, \quad (57)$$

$$R_{-+z}(\mathbf{h}, \omega_1, \omega_2, \omega_3) = 2\pi\delta(\omega_1 + \omega_2 + \omega_3) \times \left\{ \frac{1}{T} Z''(\mathbf{h}) R_0(\omega_3) K(\mathbf{h}, \omega_1) - Z'(\mathbf{h}) K(\mathbf{h}, \omega_1) K(\mathbf{h}, \omega_1 + \omega_3) \right\}.$$

Here,

$$K(\mathbf{h}, \omega) = (\omega + \mathbf{h} + i\delta)^{-1},$$

$$Z(\mathbf{h}) = \text{sh} \left[ \left( S + \frac{1}{2} \right) \frac{\mathbf{h}}{T} \right] / \text{sh} \frac{\mathbf{h}}{2T},$$

and  $R_0(\omega)$  has the same meaning as in Eq. (55), and is governed by the thermostat frequencies  $\gamma \ll \hbar$ . Averaging over  $\mathbf{y}$  is defined by Eq. (56) and in our case for large values of  $S$  we have

$$dT = \frac{1}{3} S(S+1). \quad (58)$$

However, even if  $S = 1/2$ , the deviations from Eq. (58) are small. According to Ref. 5, we have

$$dT = 1/4\sqrt{3}. \quad (59)$$

The expression (57) is the key equation. A great variety of effects can be calculated using Eq. (57). For example, the problem of slow relaxation is also related to Eq. (57), as are the preceding expressions in the present section based on Eq. (53) of Ref. 4, which is analogous to Eq. (57), in the case of a structural order-disorder glass under static conditions. Generalization to the nonequilibrium case is a relatively simple matter.

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