

Cumulant expansions and the spin-boson problem

David R. Reichman, Frank L. H. Brown, and Peter Neu

Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

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The dynamics of the dissipative two-level system at zero temperature is studied using three different cumulant expansion techniques. The relative merits and drawbacks of each technique are discussed. It is found that the noncrossing cumulant expansion technique appears to embody the virtues of the more standard cumulant methods. [S1063-651X(97)15302-8]

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I. INTRODUCTION

The standard spin-boson problem, described by the Hamiltonian [1,2]

$$H = \frac{\Delta}{2} \sigma_x + \sum_j \left[\frac{p_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 \left(x_j - \frac{c_j}{m_j \omega_j^2} \sigma_z \right)^2 \right] \quad (1)$$

has served as a paradigm for the description of dissipative effects in condensed phases. Some experimental realizations of such a Hamiltonian include, e.g., the detection of macroscopic quantum coherence in superconducting quantum interference devices [3,4], tunneling effects in metallic and insulating glasses [5], electron transfer reactions [6], and the diffusion of light interstitial particles in metals [7]. In each situation, the physical realization of the parameters in the Hamiltonian (1) is different. For instance, in metallic glasses at low temperatures the electron-hole pairs at the Fermi level constitute the bosonic bath, while for insulating glasses, tunneling effects are damped by localized and delocalized vibrational modes. Thus, the Hamiltonian (1) embodies a wealth of physical situations and has been studied in great detail (see, for instance, [1,2] and references quoted therein, or more recently [8–10]).

In order to study the dynamics of the two-level system coupled to a harmonic bath as in Eq. (1), we need a method of “tracing out” the bath or spin degrees of freedom. The bath degrees of freedom can be specified by the spectral density function,

$$J(\omega) = \frac{\pi}{2} \sum_j \frac{c_j^2}{m_j \omega_j} \delta(\omega - \omega_j), \quad (2)$$

which gives the bath density of states weighted by the square of the coupling strength between the two-level system and the bath. In most studies of the spin-boson problem, the spectral density takes the Ohmic form [1,2]

$$J(\omega) = 2\pi\alpha\omega \exp(-\omega/\omega_c), \quad (3)$$

where α is a measure of the coupling strength, and ω_c is a frequency cutoff for the bath. We note that in many cases, such as the coupling of a spin degree of freedom to a three-dimensional phonon bath in the deformation potential approximation, the spectral density (3) is not realistic, and must involve higher powers of ω .

The usual approach to finding reduced equations for the spin variables of interest involves the use of the functional integral formulation of quantum dynamics [1,2]. Formally exact equations may be found for the variables

$$P(t) = \langle \sigma_z(t) \rangle \quad (4)$$

and

$$C(t) = \frac{1}{2} \langle \{ \sigma_z(0), \sigma_z(t) \} \rangle_\beta, \quad (5)$$

where $\langle \dots \rangle_\beta$ refers to an average with respect to the canonical ensemble of Eq. (1). The quantity $P(t)$ describes the population difference in the localized spin states of the Hamiltonian (1), given that the particle is initially localized in one well and in thermal equilibrium with the bath. It is the variable of interest in certain physical situations, for example, the electron transfer problem [6]. The quantity $C(t)$, the symmetrized equilibrium correlation function of the tunneling coordinate, is related to the structure factor for neutron scattering off the tunneling particle, and is of great significance in various problems, including the antiferromagnetic Kondo problem [11]. For $C(t)$ the long-time behavior at zero temperature is known from the generalized Shiba relation, which predicts algebraic decay $C(t) \propto t^{-2}$ [2,12]. For $P(t)$ the situation is less clear, however, some studies have predicted exponential decay as $t \rightarrow \infty$ [2,13]. Despite the importance of $C(t)$, we will focus on the variable $P(t)$ in the following.

The formal path integral expression for $P(t)$ is extremely cumbersome, and a suitable approximation must be implemented to obtain useful information. The so-called “noninteracting blip approximation,” or NIBA [1,2], is the most commonly used approximation. In this scheme $P(t)$ is entirely determined by $C(t)$, i.e., $C(t) \equiv P(t)$. The NIBA may be obtained from the exact expression for $P(t)$ by invoking a series of physically based approximations. For very low temperatures, these approximations often break down, unless α is very small and only short times are considered. At zero temperature, the NIBA is not justified in the antiferromagnetic Kondo regime $\frac{1}{2} < \alpha < 1$. The NIBA also incorrectly predicts asymptotically algebraic, rather than exponential, decay for the variable $P(t)$. Lastly, NIBA incorrectly predicts that at zero temperature, $C(t) \sim t^{-2(1-\alpha)}$.

Despite these flaws, the NIBA is useful for obtaining quantitative results for $P(t)$ for high temperatures, when the

tunneling dynamics is incoherent, and in predicting the qualitative behavior of $P(t)$ for low temperatures. For instance, at zero temperature, the NIBA correctly predicts a crossover from damped oscillations to incoherent decay for the variable $P(t)$ at the point $\alpha = \frac{1}{2}$.

As shown by Aslangul *et al.* [14], the NIBA may be obtained by first applying a small polaron transformation to the Hamiltonian (1), followed by a second order application of the usual Nakijima-Zwanzig projection operator technique. It has been known for some time that this projection technique, which leads to a master equation of the convolution form, is an order by order resummation of a particular type of cumulants known as ‘‘chronologically ordered’’ cumulants [16–20]. The use of the ‘‘chronological ordering prescription,’’ or COP, when truncated at second order thus leads to the NIBA.

Interestingly, Aslangul *et al.* [15] earlier applied a convolutionless master equation technique to the study of the zero temperature spin-boson problem. This type of master equation, which can be derived by using a different type of projection operator, involves the summation of a different type of cumulant, known as ‘‘partially (time-) ordered’’ cumulants [21,17–20]. This method was probably abandoned for two reasons. First, it incorrectly describes incoherent relaxation for $P(t)$ for all values of α . Secondly, it cannot be obtained in a simple manner from the exact path integral expression. The second objection is irrelevant, since it is still possible that such an approximate resummation describes the exact behavior of $P(t)$ well. The first flaw, however, is quite serious. Despite this, the expression obtained from the ‘‘partial ordering prescription,’’ or POP, which naturally sums to an exponential form, may be expected to give a better description of $P(t)$ in the incoherent region. In fact, for values of α greater than $\frac{1}{2}$, but not too large, this method describes weakly stretched exponential relaxation, which more closely approximates the true exponential decay of $P(t)$ than does the algebraic behavior predicted by NIBA. Furthermore, as will be demonstrated in this paper, recent simulations of Egger and Mak [22] show that the POP method more accurately captures the deep decay of $P(t)$ at zero temperature for $\alpha > \frac{1}{2}$ than does the COP (NIBA) method, *even before the algebraic behavior of the NIBA is manifested.*

It is well known that by choosing a particular ordering in a truncated cumulant expansion, we are implicitly assuming different statistical properties for the relevant bath operators. The first purpose of this paper is to specify these statistical properties for the case of the spin-boson problem at zero temperature. Using this ‘‘stochastic’’ type intuition, we then discuss various cumulant ordering schemes and their associated descriptions of the behavior of $P(t)$ at $T=0$. This paper is organized as follows: In Sec. II we first present a derivation of the exact expression for $P(t)$ that allows for clear specification of the statistical properties of the bath. For this purpose, orthogonally to the conventional approach, we first integrate out the spin degrees of freedom exactly. In Sec. III, we briefly discuss the COP and POP methods. We then turn to a recently introduced cumulant method, the ‘‘noncrossing’’ cumulants [23–25]. Lastly, in Sec. IV, we compare the methods to exact simulation results.

II. MOMENT EXPANSION

We begin with an explicit expression for $P(t)$ through fourth order in Δ . We could, if we wished, obtain these terms from the exact path integral expression for $P(t)$, however, we believe that the method used in this section most clearly shows the connection to the stochastic methods upon which the cumulant expansions are based. In effect, our method offers another route to the formal expression of Refs. [1,2].

We begin with the Hamiltonian (1) in the form

$$H = H' + \frac{\Delta}{2} \sigma_x, \quad (6)$$

$$H' = \sum_k \omega_k b_k^\dagger b_k - \sigma_z \sum_k g_k (b_k^\dagger + b_k) + \sum_k \frac{g_k^2}{\omega_k}. \quad (7)$$

The quantity we wish to calculate is $P(t)$, which is defined as

$$P(t) = \langle \sigma_z(t) \rangle = Z^{-1} \text{Tr}[\exp(iHt) \sigma_z(0) \exp(-iHt) \times \pi^+ \exp(-\beta H') \pi^+], \quad (8)$$

where

$$Z = \text{Tr}[\pi^+ \exp(-\beta H') \pi^+],$$

$$\sigma_z = |L\rangle\langle L| - |R\rangle\langle R|,$$

$$\sigma_x = |L\rangle\langle R| + |R\rangle\langle L|,$$

$$\pi^+ = \frac{1}{2}(1 + \sigma_z),$$

and β is the inverse temperature. Hence, Δ is the tunneling frequency between the left ($|L\rangle$) and right ($|R\rangle$) localized states of a double-well potential. We now diagonalize Eq. (6) in the spin manifold with the use of a transformation employed by Shore and Sander [26,27] in their study of the self-trapping of an exciton coupled to phonons, namely,

$$U = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -1 \\ \phi & \phi \end{pmatrix}, \quad (9)$$

where

$$\phi = (-1)^{\sum_k b_k^\dagger b_k} = \exp\left(i\pi \sum_k b_k^\dagger b_k\right).$$

The operator ϕ is seen to be the parity operator for the bath modes. In the transformed picture, we can express

$$P(t) = -\tilde{Z}^{-1} \text{Tr}[\exp(i\tilde{H}t) \sigma_x(0) \exp(-i\tilde{H}t) \times \tilde{\pi}^+ \exp(-\beta \tilde{H}') \tilde{\pi}^+], \quad (10)$$

where

$$\tilde{H} = \frac{\Delta}{2} \phi \sigma_z + \tilde{H}', \quad (11)$$

$$\tilde{H}' = \sum_k \omega_k b_k^\dagger b_k + \sum_k g_k (b_k^\dagger + b_k) + \sum_k \frac{g_k^2}{\omega_k}, \quad (12)$$

$$\tilde{\pi}^+ = \frac{1}{2}(1 - \sigma_x), \quad (13)$$

and \tilde{Z} is now defined with respect to \tilde{H}' and $\tilde{\pi}^+$.

We now perform the trace over the spin degrees of freedom in Eq. (10), leaving

$$P(t) = \text{Re}[G(t)], \quad (14)$$

where

$$G(t) = \text{Tr}_b[\exp(iH_+t)\exp(-iH_-t)\exp(-\beta\tilde{H}')] / \text{Tr}_b[\exp(-\beta\tilde{H}')] \quad (15)$$

with

$$H_{\pm} = \pm \frac{\Delta}{2} \phi + \tilde{H}'.$$

This trace over the bath degrees of freedom is most easily performed in the small polaron representation, defined by the transformation

$$U = \exp(\xi), \quad (16)$$

$$\xi = \sum_k \frac{g_k}{\omega_k} (b_k - b_k^\dagger). \quad (17)$$

In this picture, we may express $G(t)$ as

$$\left\langle \exp_{-} \left(i \int_0^t d\tau \eta(\tau) \right) \exp_{-} \left(i \int_0^t d\tau \eta(\tau) \right) \right\rangle_B, \quad (18)$$

where

$$\eta(t) = \frac{\Delta}{2} \exp[-\xi(t)] \phi \exp[\xi(t)], \quad (19)$$

and

$$\xi(t) = \sum_k \frac{g_k}{\omega_k} (b_k e^{-i\omega_k t} - b_k^\dagger e^{i\omega_k t}). \quad (20)$$

The averaging (denoted by $\langle \dots \rangle_B$) is over the canonical ensemble of harmonic oscillators, [i.e., $\rho_B = \exp(-\beta \sum_k \omega_k b_k^\dagger b_k) / \text{Tr}_b \exp(-\beta \sum_k \omega_k b_k^\dagger b_k)$], and \exp_{-} (\exp_{+}) denotes a time ordered exponential with latest time to the right (left). From this point on, all averaging will be with respect to this ensemble, and we will drop the subscript B . Since the spin degree of freedom has been removed, our method allows us to focus on the bath operators that arise in the expansion of $P(t)$. Using the following properties of the parity operator,

$$\phi \exp[\xi(t)] = \exp[-\xi(t)] \phi$$

and

$$\phi^2 = 1,$$

we can show, through fourth order in Δ , the moment expansion for $P(t)$,

$$P(t) = 1 + \int_0^t dt_1 \int_0^{t_1} dt_2 m_2(t_1 t_2) + \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 m_4(t_1, t_2, t_3, t_4) + \dots, \quad (21)$$

where the moments m_i equal

$$m_2(t_1, t_2) = -\Delta^2 \text{Re} \langle B_{-}(t_1) B_{+}(t_2) \rangle, \quad (22)$$

$$m_4(t_1, t_2, t_3, t_4) = \frac{\Delta^4}{4} \text{Re} \left[\langle B_{-}(t_1) B_{+}(t_2) B_{-}(t_3) B_{+}(t_4) \rangle + \langle B_{-}(t_2) B_{+}(t_1) B_{-}(t_3) B_{+}(t_4) \rangle + \langle B_{-}(t_3) B_{+}(t_1) B_{-}(t_2) B_{+}(t_4) \rangle + \langle B_{-}(t_4) B_{+}(t_1) B_{-}(t_2) B_{+}(t_3) \rangle \right], \quad (23)$$

and

$$B_{\pm}(t) = \exp[\pm 2\xi(t)]. \quad (24)$$

Note that $m_{2n-1} = 0$. In this paper, we shall only use the first two nonvanishing moments, although it is a simple matter to execute the expansion to an arbitrary order. From (21)–(23) we conclude that $P(t)$ is entirely determined by the statistical properties of the bath operators $B_{\pm}(t)$ with respect to the canonical state of the bath. Note that the operators B_{\pm} always appear in pairs. In order to specify the statistics obeyed by the operators B_{\pm} , we now calculate the second and fourth moment of the B_{\pm} 's. It is a simple matter to show that

$$\langle B_{-}(t_1) B_{+}(t_2) \rangle = \exp[-iQ_1(t_1 - t_2) - Q_2(t_1 - t_2)], \quad (25)$$

where

$$Q_1(t_1 - t_2) = 4 \sum_k \left(\frac{g_k}{\omega_k} \right)^2 \sin[\omega_k(t_1 - t_2)], \quad (26)$$

and

$$Q_2(t_1 - t_2) = 4 \sum_k \left(\frac{g_k}{\omega_k} \right)^2 \{1 - \cos[\omega_k(t_1 - t_2)]\} \coth(\beta\omega_k/2). \quad (27)$$

Furthermore, by using the relation $e^{A+B} = e^A e^B e^{-1/2[A,B]}$ and $\langle e^{\kappa b^\dagger} e^{-\kappa^* b} \rangle = e^{-|\kappa|^2}$ for $[A, B]$, κ being c numbers, one can also show that

$$\langle B_{-}(t_1) B_{+}(t_2) B_{-}(t_3) B_{+}(t_4) \rangle = \frac{\langle B_{-}(t_1) B_{+}(t_2) \rangle \langle B_{-}(t_3) B_{+}(t_4) \rangle \langle B_{-}(t_1) B_{+}(t_4) \rangle \langle B_{-}(t_2) B_{+}(t_3) \rangle}{\langle B_{-}(t_1) B_{+}(t_3) \rangle \langle B_{-}(t_2) B_{+}(t_4) \rangle}. \quad (28)$$

This property can be extended to an arbitrary number of B_{\pm} pairs. This gives a type of ‘‘Wick’’ theorem for the operators B_{\pm} , and demonstrates the underlying reason why only the functions Q_1 and Q_2 appear in the exact path integral [see Eqs. (4)–(17) to (4)–(22) in Ref. [1]]. It can now be explicitly checked that the expression (21) is identical to the exact path integral expression, at least through the fourth moment. Note that the property (28) is different from the statistical properties held by commonly used stochastic processes such as Gaussian, two-state-jump, or Gaussian random matrix processes. We will return to this point in the next section.

The moment expansion itself is not a very useful scheme for describing dynamics, because an arbitrary truncation of the expansion leads to secular terms that grow with time. We next resort to schemes that provide partial (approximate) resummations of the moment expansion to all orders. Such schemes are the cumulant expansions that will be introduced in the next section.

III. CUMULANT EXPANSIONS

We now discuss the various ordering prescriptions that allow for partial resummation of the expansion (21). Each ordering method leads to a unique type of master equation [24]. We note that, when carried out to infinite order, all of the ordering techniques give the same (exact) result. When truncated at a finite order, however, the results are different. In simple stochastic situations, when the temperature of the bath is infinite and the generator for time evolution (the Liouville operator) commutes with itself for all times, i.e., $[L(t), L(t')] = 0$, the use of a particular truncated cumulant expansion implies a knowledge of the stochastic properties of the bath functions. In simple cases, truncation of the cumulant expansion in the ‘‘correct’’ ordering prescription can lead to *exact* results that may be obtained in the ‘‘incorrect’’ ordering prescription only at infinite order. In the quantum case described by the Hamiltonian (1), where $[L(t), L(t')] \neq 0$, truncation of a cumulant expansion at finite order in any ordering prescription will never lead to exact results due to the noncommutivity of the Liouvillian at different times [28–30]. It is precisely this noncommutivity that leads to the variety of time orderings of the operators B_{\pm} in the expression (23) for m_4 . Despite this fact, the statistical properties of the bath operators still dictate the choice of ordering prescription that provides the most rapid convergence of the cumulant series (if such convergence exists) [31].

We begin by discussing the chronological ordering prescription, or COP. In this prescription, a master equation of the form (see, for instance, [20,17–19,24])

$$\frac{dP(t)}{dt} = \int_0^t K^{\text{COP}}(t, \tau) P(\tau) d\tau \quad (29)$$

is obtained. This equation may be expressed in the form

$$\begin{aligned} \frac{dP(t)}{dt} = & \sum_{n=2}^{\infty} \int_0^t d\tau_1 \cdots \int_0^{\tau_{n-2}} d\tau_{n-1} \\ & \times \gamma_n(t, \tau_1, \dots, \tau_{n-1}) P(\tau_{n-1}). \end{aligned} \quad (30)$$

The COP cumulants γ are obtained from the moments by a recursion relation [24]. In the present case this yields

$$\begin{aligned} \gamma_{2n-1} &= 0, \\ \gamma_2(t, \tau_1) &= m_2(t, \tau_1), \\ \gamma_4(t, \tau_1, \tau_2, \tau_3) &= m_4(t, \tau_1, \tau_2, \tau_3) \\ &\quad - m_2(t, \tau_1) m_2(\tau_2, \tau_3), \dots \end{aligned} \quad (31)$$

For the simple case where the stochastic Liouvillian commutes with itself for all times, all of the COP cumulants γ_n vanish for $n \geq 3$ if the stochastic bath functions have the two-state-jump behavior [20]

$$\begin{aligned} \langle B(t_1) B(t_2) B(t_3) B(t_4) \cdots \rangle &= \langle B(t_1) B(t_2) \rangle \\ &\quad \times \langle B(t_3) B(t_4) \cdots \rangle, \end{aligned} \quad (32)$$

for $t_1 > t_2 > t_3 > t_4 > \cdots$, where $B(t)$ is the stochastic bath function responsible for system dissipation. If these bath functions have different statistics, it may not be a good approximation to truncate the series at low orders.

Returning now to the quantum case of interest in this paper, we find at lowest order, as shown by Aslangul *et al.* [14], the NIBA equation for $P(t)$:

$$\frac{dP(t)}{dt} = \int_0^t m_2(t-\tau) P(\tau) d\tau, \quad (33)$$

where, at $T=0$, using the Ohmic constraint (3), along with Eq. (22), we may express

$$m_2(t-\tau) = -\Delta^2 \text{Re} \frac{1}{[1 + i\omega_c(t-\tau)]^{2\alpha}}. \quad (34)$$

As shown by Grabert and Weiss [32], the solution to Eq. (33) with the kernel (34) can be given for all $\alpha < 1$ (in the limit $\Delta/\omega_c \rightarrow 0$) by the Mittag-Leffler function [33],

$$P_{\text{NIBA}}(y) = E_{2(1-\alpha)}(-y^{2(1-\alpha)}), \quad (35)$$

where $y = \Delta_{\text{eff}} t$ and

$$\Delta_{\text{eff}} = \Delta [\cos(\pi\alpha) \Gamma(1-2\alpha)]^{1/2(1-\alpha)} \left(\frac{\Delta}{\omega_c} \right)^{\alpha/(1-\alpha)}. \quad (36)$$

This solution shows damped oscillations for $\alpha < \frac{1}{2}$, and incoherent decay for $\alpha \geq \frac{1}{2}$. This behavior has been qualitatively confirmed by Monte Carlo simulation [22]. As mentioned in the Introduction, the NIBA cannot give the correct asymptotic decay of $P(t)$ [yielding the algebraic decay $P(t) \propto t^{-2(1-\alpha)}$ rather than exponential decay], and is unable to account for the depth of the decay in the region $\alpha \geq \frac{1}{2}$ even before the incorrect algebraic behavior sets in. The NIBA is, however, known to work quite well for short times and weak coupling strengths. The analysis given in the last section provides an explanation for this fact. For ‘‘short’’ times and ‘‘small’’ values of α the function $m_2(t)$ is a rather broad, weakly decaying function of time. When this is the case, the statistical property (28) of the operators B_{\pm} is approximately of the two-state-jump form (32) *as far as the integrations over the cumulants $\gamma_{n \geq 3}$ are concerned*. This approximate

equivalence holds in a *stochastic* sense, in that all of the four point correlation functions in m_4 [see Eq. (23)] may be approximated by $m_2(t_1, t_2)m_2(t_3, t_4)$. For such times and coupling strengths, the NIBA will be essentially exact, as all COP cumulants for $n \geq 3$ will vanish when integrated. We shall not provide precise meaning to the terms ‘‘short’’ or ‘‘small,’’ although their meaning should be clear in the context of the present discussion, and could be quantified without undue labor [in fact ‘‘short’’ and ‘‘small’’ will be coupled in the sense that the effective time scale of oscillation or decay, $(\Delta_{\text{eff}})^{-1}$, depends on α]. Note that the statistical property (28) trivially gives two-state-jump behavior for $\alpha=0$, which leads to the correct behavior $P(t)=\cos(\Delta t)$. While this is obvious, other cumulant techniques (such as those discussed below) do not embody this type of statistics for $\alpha=0$, and cannot give the correct, freely oscillating solution for zero coupling strength upon truncation at second order. The statement (often given in the literature [34]) that NIBA works for weak coupling because it is a perturbative scheme is thus not strictly correct.

The (somewhat heuristically) demonstrated fact that the property (28) can resemble two-state-jump behavior under certain circumstances leads one to believe that extending the COP scheme to fourth order would not be useful, since this property is reflected in the vanishing of all COP cumulants higher than the second. Extending the COP method to fourth order *does not give a method for computing ‘‘interblip’’ interactions* in the language of Ref. [1].

We now turn to the partial ordering prescription, or POP. At second order, this method was applied by Aslangul *et al.* [15] to the spin-boson problem at $T=0$. The POP master equation has a convolutionless [20,24] form

$$\frac{dP(t)}{dt} = \left(\int_0^t K^{\text{POP}}(\tau) d\tau \right) P(t). \quad (37)$$

$K^{\text{POP}}(t)$ may be obtained from the moments

$$K^{\text{POP}}(t) = \sum_{n=1}^{\infty} \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \cdots \int_0^{\tau_{n-1}} d\tau_n \times \theta_{n+1}(t, \tau_1, \dots, \tau_n), \quad (38)$$

where

$$\begin{aligned} \theta_{2n-1} &= 0, \\ \theta_2(t, \tau_1) &= m_2(t, \tau_1), \\ \theta_4(t, \tau_1, \tau_2, \tau_3) &= m_4(t, \tau_1, \tau_2, \tau_3) - m_2(t, \tau_1)m_2(\tau_2, \tau_3) \\ &\quad - m_2(t, \tau_2)m_2(\tau_1, \tau_3) \\ &\quad - m_2(t, \tau_3)m_2(\tau_1, \tau_2), \dots \end{aligned} \quad (39)$$

The POP resummation is exact at second order for the simple case of a classical Gaussian stochastic process. We note that the statistical property (28) appears to be very different from the standard Wick theorem for Gaussian processes. We may still expect that the POP method is better suited for the incoherent regime $\alpha \geq \frac{1}{2}$ for the following reasons. First, the POP technique resums to an exponential form, which is expected to better capture the long time behavior of $P(t)$, which is expected to be exponential. In general, the POP method sums (infinitely) more terms than the COP method does. For example, expansion of the second order truncation in the COP gives to fourth order

$$P(t) = 1 + \int_0^t dt_1 \int_0^{t_1} dt_2 m_2(t_1, t_2) + \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 m_2(t_1, t_2) m_2(t_3, t_4) + \cdots,$$

whereas the POP gives

$$\begin{aligned} P(t) &= 1 + \int_0^t dt_1 \int_0^{t_1} dt_2 m_2(t_1, t_2) + \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 m_2(t_1, t_2) m_2(t_3, t_4) \\ &\quad + m_2(t_1, t_4) m_2(t_2, t_3) + \cdots \end{aligned}$$

Clearly, the extra terms do not ensure a more accurate result. For example, for weak coupling strengths, the POP must be carried out to infinite order to obtain coherent behavior. However, in the incoherent regime, the effective time scale, defined by Eq. (36) is very long, while the decay of the function $m_2(t)$ is ‘‘slow’’ (algebraic). In this case, we may expect that we must include terms such as $m_2(t_1, t_4)m_2(t_2, t_3)$ that extend over large portions of the integration region. As we will show in the next section, the POP method seems to capture the behavior of $P(t)$ better than the COP method in the incoherent regime, even before the full asymptotic behavior is displayed.

At second order [$K^{\text{POP}}(t, \tau) = m_2(t - \tau)$] the POP equation (38) may be solved [15]:

$$\begin{aligned} P(t) &= \exp \left[\frac{\Delta^2}{4\omega_c^2} \frac{1}{(\alpha - 1/2)(1 - \alpha)} \right. \\ &\quad \left. \times \left(1 - \frac{\cos(2(1 - \alpha)\tan^{-1}\omega_c t)}{(1 + \omega_c^2 t^2)^{\alpha-1}} \right) \right]. \end{aligned} \quad (40)$$

Note that Eq. (40) describes a *stretched exponential* rather than exponential decay. For values of α that are not too much larger than $\frac{1}{2}$, however, the POP expression should give a better representation of the asymptotic exponential decay of $P(t)$ than the COP (NIBA) expression, which yields algebraic decay asymptotically.

We have now given some motivation for the belief that

the COP method (at lowest order) should give a better description of $P(t)$ in the region $\alpha < \frac{1}{2}$ while the POP method should be better in the incoherent region $\alpha \geq \frac{1}{2}$. We now ask whether there is a summation method that is a ‘‘hybrid’’ of the two methods, in the sense that it can incorporate at low order features of the COP and POP methods. In the theory of stochastic processes, such a technique has recently been developed [23–25]. This method is based on the summation of ‘‘noncrossing’’ (NC) cumulants (for a precise definition see Refs. [23–25]). For simple stochastic situations, if the coupling is not too strong, the NC technique (including terms up to fourth order) has been shown to interpolate between the two-state-jump behavior and the Gaussian behavior [24].

The NC description leads to a *nonlinear* equation of motion for $P(t)$ [24], which at second order, may be expressed as

$$\frac{dP(t)}{dt} = M(t), \quad (41)$$

where

$$M(t) = \int_0^t dt_1 \zeta_2(t-t_1) P(t-t_1) P(t_1). \quad (42)$$

To fourth order the master equation for $P(t)$ in the NC scheme reads

$$\begin{aligned} \frac{dP(t)}{dt} = & M(t) + \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \zeta_4(t, t_1, t_2, t_3) \\ & \times P(t-t_1) P(t_1-t_2) P(t_2-t_3) P(t_3). \end{aligned} \quad (43)$$

As in the previous two case, the NC cumulants ζ may be obtained from the moments by a recursion relation [23,25]. In the present case this yields up to fourth order

$$\begin{aligned} \zeta_{2n-1} &= 0, \\ \zeta_2(t, t_1) &= m_2(t, t_1), \\ \zeta_4(t, t_1, t_2, t_3) &= m_4(t, t_1, t_2, t_3) - m_2(t, t_1) m_2(t_2, t_3) \\ &\quad - m_2(t, t_3) m_2(t_1, t_2), \dots \end{aligned} \quad (44)$$

It is clear that in appearance, the NC cumulants are a ‘‘compromise’’ between the COP and POP cumulants. We note in passing two interesting facts. First, in the stochastic realm, the NC ordering prescription truncated at second order is exact for the case of a stochastic bath modeled by symmetric ($N \times N$) Gaussian random matrices for the commutator of $\eta(t)$'s. In this case, the ‘‘crossing contraction’’ $m_2(t, t_2) m_2(t_1, t_3)$ vanishes by means of a $1/N$ argument for $N \rightarrow \infty$. This leads naturally to the equation (42) and its systematic generalization (43) through the NC cumulants. Equation (42) has first been derived by Kraichnan [36–39] in the fields of turbulence and fluid dynamics. Our motivation for the application of this method is *not* based on a stochastic type of reasoning, but on the fact that in simple situations this ordering prescription may combine the benefits of the COP and POP methods.

Before concluding this section, we would like to apply all three ordering prescriptions to the case $T=0$, $\alpha = \frac{1}{2}$. Here, it is known that the ‘‘exact’’ (in the sense specified in Ref. [1]) result for $P(t)$ is

$$P(t) = \exp\left[-\frac{\pi}{2} \omega_c \left(\frac{\Delta}{\omega_c}\right)^2\right] \quad (45)$$

in the limit $\Delta/\omega_c \rightarrow 0$. Note that in this limit, the second moment becomes δ correlated ($t \geq 0$)

$$m_2(t) \rightarrow \frac{\pi \Delta^2}{2 \omega_c} \delta(t).$$

Using the fact that $P(0) = 1$, it is clear that *all three ordering prescriptions give the same result* given by Eq. (45) at second order. Hence, the value $\alpha = \frac{1}{2}$ corresponds to the *white noise* limit of the bath operators $B_{\pm}(t)$.

IV. RESULTS AND CONCLUSIONS

Before comparing the results of the three ordering methods, we make some comments on the methods discussed in Sec. III. We have shown how three different cumulant methods give rise to different master equations with different properties. We have tried to physically motivate when each approach should have success when applied to the spin-boson problem at zero temperature. Note that in general, the discussion of convergence of each cumulant series is a difficult task. This task is made more difficult by the fact that, at zero temperature, the algebraic decays of the bath correlation functions leave us with no clearly defined relaxation time for the bath. This means that we will rely almost exclusively on physical considerations and comparison with accepted results to determine the success or failure of the methods employed. The case of finite temperature, which can be studied by the same methods employed here, is often easier in this respect. If an exponential correlation time τ_b can be assigned to the decaying bath correlation functions, then it is possible to consider a systematic expansion in $\Delta_{\text{eff}} \tau_b$ provided that this dimensionless parameter is small. When this is the case, the POP provides the most facile way of systematically summing terms in the parameter $\Delta_{\text{eff}} \tau_b$ [21]. In case of Ohmic dissipation and finite temperature T , the characteristic correlation time of the $B_{\pm}(t)$ is given by $\tau_b \sim (2\pi\alpha T)^{-1}$ [1]. This point of view provides an explanation for the familiar statement that the NIBA works well in the incoherent tunneling regime $\Delta_{\text{eff}} \tau_b \ll 1$. In a stochastic language, this parameter region corresponds to the *narrowing* or *Markov* limit of the B_{\pm} 's. Similarly to the white noise limit mentioned above, one finds that all three cumulants schemes work well already at second order and provide essentially the same behavior, $P(t) \approx \exp[-\Delta_{\text{eff}}^2(T) \tau_b t]$ with $\Delta_{\text{eff}}(T) \propto T^\alpha$ [1].

Since we expect the NIBA to be accurate for very weak coupling strengths, we first turn to the case of weak to intermediate coupling strength, $\alpha = 0.3$. For coupling strengths in this range, simulations at low temperatures have shown that the NIBA is qualitatively correct in predicting damped oscillations, but may fail in predicting the damping strength. An example of this is given by the simulations of Makarov and Makri [13], which show that for intermediate coupling, NIBA may fail by slightly underestimating the number of

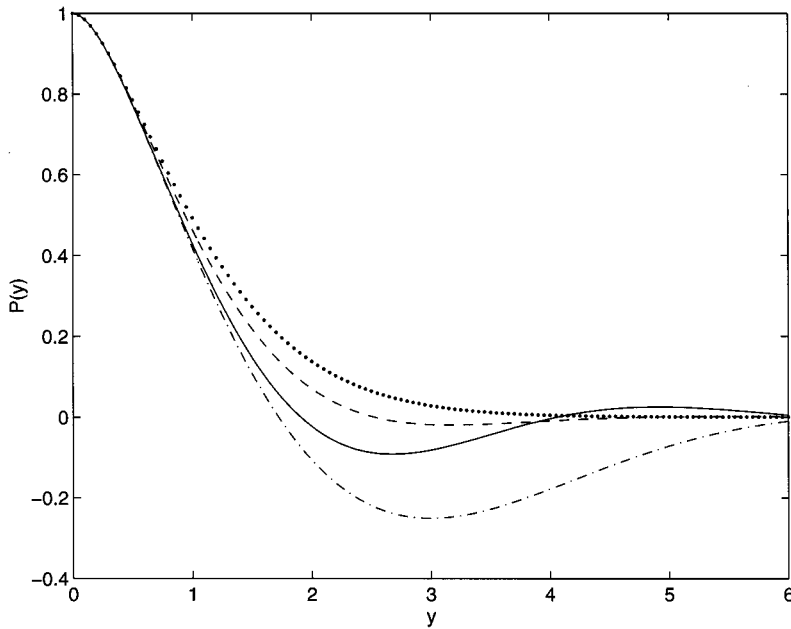


FIG. 1. Zero temperature plot of $P(y)$ ($y = \Delta_{\text{eff}} t$) for $\alpha = 0.3$ and $\omega_c/\Delta = 6$. The dotted line is the second order POP result, the dashed line is the second order noncrossing cumulant result, the dash-dotted line is the NIBA (second order COP) result, and the solid line is the fourth order noncrossing cumulant result.

oscillations in $P(t)$. We note, however, that these simulations were carried out for values of Δ/ω_c that are not very small. In Fig. 1, we plot the NIBA (second order COP) solution for $P(t)$ against the solutions obtained from second and fourth order truncations of the noncrossing cumulant method, and the second order POP. Note that, as expected, the second order POP solution for $P(t)$ fails to produce any oscillations. We expect that for $\alpha < \frac{1}{2}$ the POP will always be inaccurate at low orders. The second order noncrossing cumulant solution for $P(t)$, obtained from the Kraichnan-type equation (41)–(42) is similar to the NIBA solution, although the oscillation in $P(t)$ is much weaker. The fourth order noncrossing cumulant solution gives a first oscillation that is very similar in magnitude to the NIBA solution, however, it describes one extra weak remnant of an oscillation. This behavior is very similar to the behavior displayed in the exact

simulations of Makarov and Makri [13]. Although this example represents only one value of α , similar results may be obtained for all moderately strong values of α up to $\alpha = \frac{1}{2}$. Thus, it appears that the noncrossing scheme works well in incorporating (and perhaps improving) the qualities of the COP method for moderate values of α when $\alpha < \frac{1}{2}$.

We now turn to the relaxation of $P(t)$ in the incoherent regime $\alpha \geq \frac{1}{2}$. Here, the beautiful path integral simulations of Egger and Mak [22] provide a means of comparing the cumulant expansion methods with exact results. In this region, we expect the POP to be most successful, while the NIBA (second order COP) is expected to be worse. Based on experience with simple stochastic situations, we hope, as in the coherent portion of the coupling space, that the noncrossing scheme can capture the essence of the POP in this regime. In Fig. 2, we show the decay of $P(t)$ calculated by different

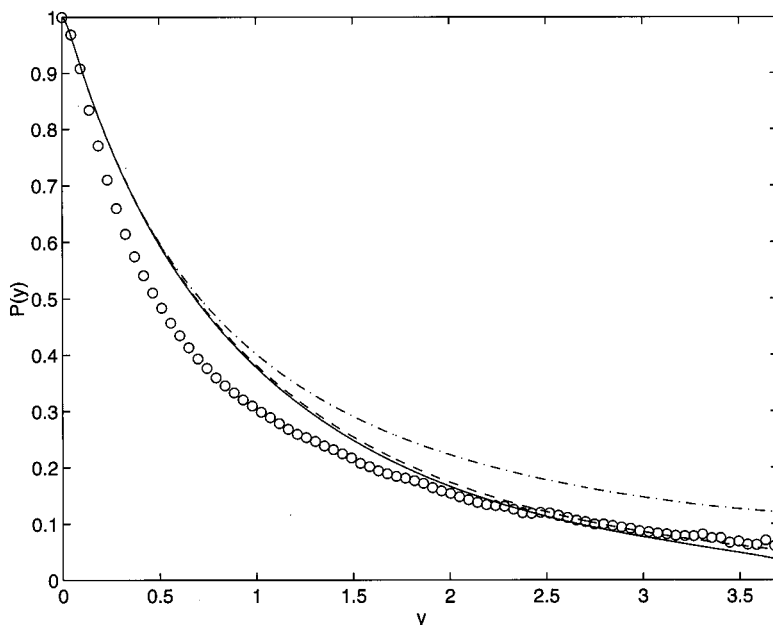


FIG. 2. Zero temperature plot of $P(y)$ ($y = \Delta_{\text{eff}} t$) for $\alpha = 0.6$ and $\omega_c/\Delta = 6$. The dash-dotted line is the NIBA (second order COP) result, the dashed line is the second order POP result, the solid line is the second order noncrossing cumulant result, and the open circles are the simulation result of Egger and Mak [22].

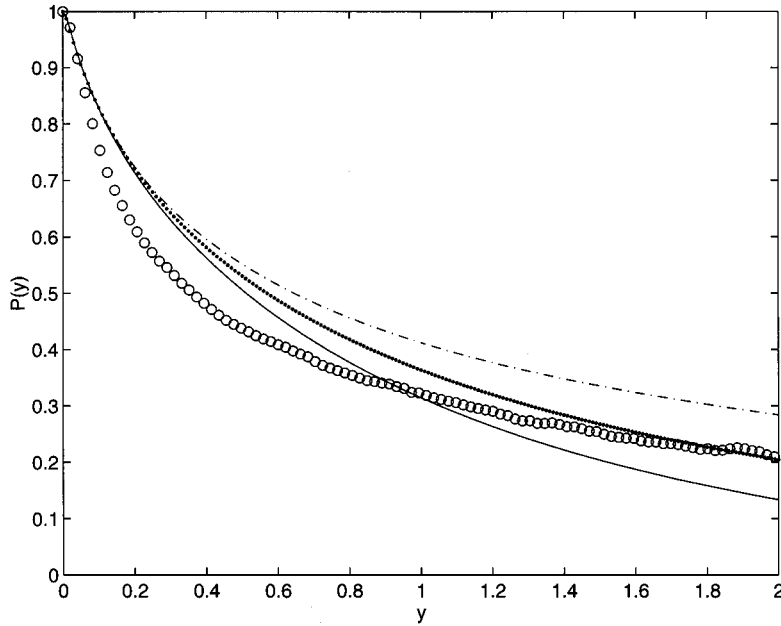


FIG. 3. Zero temperature plot of $P(y)$ ($y = \Delta_{\text{eff}} t$) for $\alpha = 0.7$ and $\omega_c / \Delta = 6$. The dash-dotted line is the NIBA (second order COP) result, the dotted line is the second order POP result, the solid line is the second order noncrossing cumulant result, and the open circles are the simulation result of Egger and Mak [22].

ordering prescriptions for $\alpha = 0.6$. It must be noted that the simulations were carried out for long times, but not long enough to show the asymptotic algebraic decay of the NIBA (second order COP) solution of $P(t)$, or the asymptotic exponential decay of the exact solution. Due to this fact, it is somewhat difficult to see that the POP solution is to be preferred over the NIBA solution in the incoherent regime. In support of this claim we note two facts. First, the POP solution better manifests the deep decay of the exact result in the nonasymptotic regime. Secondly, near the point $\alpha = 1/2$, the POP solution will better approximate the exponential decay of $P(t)$ in the asymptotic regime than the NIBA, which predicts $P(t) \sim t^{-2(1-\alpha)}$. The simulations of Egger and Mak end just before this regime is reached. As we had hoped, for $\alpha = 0.6$, the second order noncrossing technique is nearly identical to the POP. Figure 3 shows the results for $\alpha = 0.7$.

Again, the POP seems to perform the best, while the second order noncrossing scheme overestimates the decay. As in the case of weaker coupling, we see if truncation after fourth order in the noncrossing cumulants can properly correct the second order result. This test is shown in Fig. 4. While the results appear to show that the noncrossing scheme is converging to a POP-like description of $P(t)$, we again must exercise caution due to the lack of further concrete evidence for this belief. For such a large value of α , it is quite possible that the cumulant methods break down.

One interesting property displayed in Figs. 2 and 3 is the close agreement between the POP description of $P(t)$ and the exact simulation of $P(t)$ for moderately long times. In Fig. 3 this behavior occurs for $y \geq 2.5$, while in Fig. 3, the agreement is less pronounced, but appears to occur for $y \geq 1.6$ (note that there is some scattering in the simulation

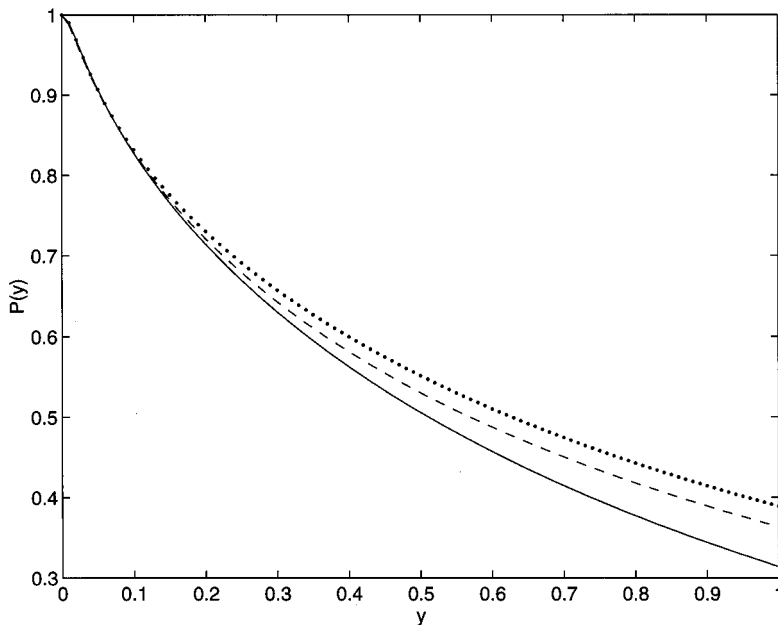


FIG. 4. Zero temperature plot of $P(y)$ ($y = \Delta_{\text{eff}} t$) for $\alpha = 0.7$ and $\omega_c / \Delta = 6$. The dotted line is the fourth order noncrossing cumulant result, the dashed line is the second order POP result, and the solid line is the second order noncrossing cumulant result. Note the change in the x axis.

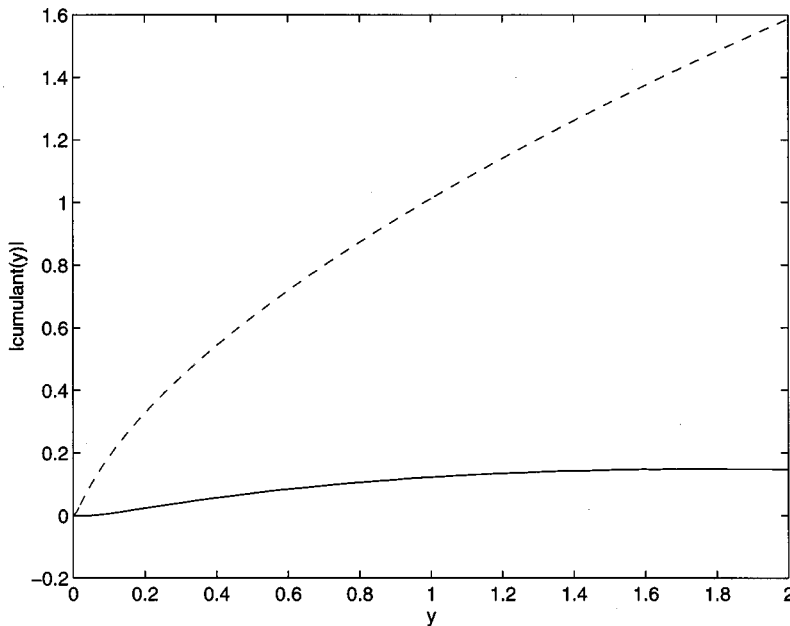


FIG. 5. Relative magnitude of second and fourth cumulant effects in the POP for $\alpha=0.7$. The dashed line shows $|\int_0^y dy' \int_0^{y'} dy'' K_2^{\text{POP}}(y'')|$ and the solid line shows $|\int_0^y dy' \int_0^{y'} dy'' K_4^{\text{POP}}(y'')|$. $K_n^{\text{POP}}(t)$ is defined in Eq. (38).

data for $\alpha=0.7$ near $y=2$ that slightly obscures the apparent agreement). In order to investigate this, we have computed $\int_0^y dy' \int_0^{y'} dy'' K_2^{\text{POP}}(y'')$ and $\int_0^y dy' \int_0^{y'} dy'' K_4^{\text{POP}}(y'')$ where $K_n^{\text{POP}}(t)$ is the n th term in the expansion (38). If the integrated second order POP cumulant is of order one for a given time interval, while all other POP cumulants are small when integrated over the appropriate time domain, then we expect the truncation at second order to be a good approximation. While we cannot study all the POP cumulants, we have studied the second and the fourth. In Fig. 5, we compare the properties of the second and fourth POP cumulants for $\alpha=0.7$. For $y=1.4$ to $y=2$ (the boundary of the simulation results of Egger and Mak [22]), we see that the contribution from the second POP cumulant is at least ten times greater than the contribution from the fourth cumulant. This strongly suggests that the agreement of the second order POP method with the exact simulations is no coincidence. In fact, the agreement between Eq. (41) and the simulation occurs precisely in the interval where the second order cumulant dominates the fourth order cumulant. Since the slopes of the two curves suggest that this behavior continues for some time, we feel there is strong evidence for the somewhat remarkable conclusion that, for significant intermediate times, the decay of $P(t)$ is quantitatively described by a stretched exponential. For longer times, the decay is most likely purely exponential.

We now summarize the results presented in this paper. We first carried out a derivation of the moment expansion for the variable $P(t)$ in the spin-boson problem. We then used

this derivation to discuss the “statistical” properties of the relevant bath operators. Using the moment expansion, we first discussed the chronological and partial ordering prescriptions that involve different types of cumulants. We discussed the merits and drawbacks of each method. In an effort to combine the merits of the COP and the POP, we applied the noncrossing scheme. Specializing to the case of zero temperature, we tested each method, including fourth order terms when necessary. Our results show that the noncrossing scheme is a promising candidate for combining the virtues of the COP and POP, especially for intermediate values of α on either side of the coherent-incoherent transition value of $\alpha=\frac{1}{2}$. We note that more work should be done to test the validity of this claim. Lastly, we have provided evidence to support the belief that the stretched exponential behavior described by second order truncation of the POP in the incoherent portion of coupling space may in fact be very accurate for intermediate times.

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[1] A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg, and W. Zwerger, *Rev. Mod. Phys.* **59**, 1 (1987).

[2] U. Weiss, *Quantum Dissipative Systems*, Series in Modern Condensed Matter Physics Vol. 2 (World Scientific, Singapore, 1993).

[3] A. J. Leggett, in *Percolation, Localization and Superconductivity*, edited by A. Goldmann and S. A. Wolf, Vol. 109 of *NATO Advanced Studies Institute Series B: Physics* (Plenum, New York, 1984), p. 1.

[4] L. Esaki, in *Highlights in Condensed Matter Physics and Fu-*

- ture Prospects, edited by L. Esaki (Plenum, New York, 1991), p. 55.
- [5] For a review, see *Amorphous Solids – Low Temperature Properties*, Topics in Current Physics Vol. 24, edited by W. A. Phillips (Springer, Berlin, 1984).
- [6] R. Marcus, *J. Chem. Phys.* **24**, 966 (1956); A. Garg, J. N. Onuchi, and V. Ambegaokar, *ibid.* **83**, 4491 (1985). For a review see R. A. Marcus and N. Sutin, *Biochim. Biophys. Acta* **811**, 265 (1985); G. R. Fleming and P. G. Wolynes, *Phys. Today* **5**, 36 (1990).
- [7] H. Grabert and H. R. Schober, *Hydrogen in Metals III*, edited by H. Wipf (Springer, Heidelberg, 1996). For a review see *Philos. Trans. R. Soc. London A* **350**, 169 (1995).
- [8] S. K. Kehrein, A. Mielke, and P. Neu, *Z. Phys. B* **99**, 269 (1996).
- [9] S. K. Kehrein and A. Mielke, *Ann. Phys. (Leipzig)* (to be published).
- [10] A. Würger, in *From Coherent Tunneling to Relaxation, Dissipative Quantum Dynamics of Interacting Defects*, Springer Tracts in Modern Physics Vol. 135 (Springer, Berlin, 1997).
- [11] A. W. W. Ludwig and I. Affleck, *Nucl. Phys.* **B428**, 545 (1994).
- [12] M. Sasseti and U. Weiss, *Phys. Rev. Lett.* **65**, 2262 (1990).
- [13] N. Makri and D. E. Makarov, *J. Chem. Phys.* **102**, 4600 (1995).
- [14] C. Aslangul, N. Pottier, and D. Saint-James, *J. Phys. (Paris)* **47**, 1657 (1986).
- [15] C. Aslangul, N. Pottier, and D. Saint-James, *J. Phys. (Paris)* **46**, 2031 (1985).
- [16] R. H. Terwiel, *Physica* **74**, 248 (1974).
- [17] B. Yoon, J. M. Deutsch, and J. H. Freed, *J. Chem. Phys.* **62**, 4687 (1975).
- [18] S. Mukamel, I. Oppenheim, and J. Ross, *Phys. Rev. A* **17**, 1988 (1978).
- [19] S. Mukamel, *Chem. Phys.* **37**, 33 (1978).
- [20] F. Shibata and T. Arimitsu, *J. Phys. Soc. Jpn.* **49**, 891 (1980).
- [21] N. G. van Kampen, *Physica* **74**, 215 (1974); **74**, 239 (1974).
- [22] R. Egger and C. H. Mak, *Phys. Rev. B* **50**, 15 210 (1994).
- [23] R. Speicher, *Math. Ann.* **298**, 611 (1994).
- [24] P. Neu and R. Speicher, *Z. Phys. B* **92**, 399 (1993).
- [25] P. Neu and R. Speicher, *J. Stat. Phys.* **80**, 1279 (1995).
- [26] H. B. Shore and L. M. Sander, *Phys. Rev. B* **7**, 4537 (1973).
- [27] M. Wagner, *Unitary Transformation in Solid State Physics*, Modern Problems in Condensed Matter Science Vol. 15 (North-Holland, Amsterdam, 1986).
- [28] R. F. Fox, *J. Math. Phys.* **17**, 1148 (1976).
- [29] I. B. Rips and V. Capek, *Phys. Status Solidi B* **100**, 451 (1980).
- [30] J. Budimir and J. L. Skinner, *J. Stat. Phys.* **49**, 1029 (1987).
- [31] M. Aihara, H. M. Sevian, and J. L. Skinner, *Phys. Rev. A* **41**, 6596 (1990).
- [32] H. Grabert and U. Weiss, *Phys. Rev. Lett.* **54**, 1605 (1985).
- [33] A. Erdelyi, *Higher Transcendental Functions* (McGraw-Hill, New York, 1955), Vol. 3.
- [34] G. Stock, *Phys. Rev. E* **51**, 3038 (1995).
- [35] D. E. Makarov and N. Makri, *Chem. Phys. Lett.* **221**, 482 (1994).
- [36] R. H. Kraichnan, *J. Math. Phys.* **2**, 124 (1961).
- [37] U. Frisch and R. Bourret, *J. Math. Phys.* **11**, 364 (1970).
- [38] R. H. Kraichnan, *Phys. Rev.* **109**, 1407 (1958); *J. Fluid. Mech.* **5**, 497 (1959).
- [39] R. H. Kraichnan, *J. Math. Phys.* **3**, 475 (1962); **3**, 496 (1962).