

Asymptotic regime of quantal stochastic and dissipative motion

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We analyze the irreversible behavior of a quantum harmonic oscillator immersed in an arbitrary thermal reservoir. For the fully coupled (FC) model, i.e., the coupling between subsystems linear in the coordinate of the oscillator, we construct a generalized master equation for the reduced density operator of the oscillator. We apply the rotating-wave approximation (RWA) and investigate its connection with the FC master equation. By means of the Wigner distribution, we obtain the corresponding Fokker-Planck equations in the semiclassical representation. We find that, when the RWA is not imposed, the correct classical limit is obtained (Kramers equation). Then, we establish the conditions for the equivalence between the FC Fokker-Planck equation and that obtained in the RWA. Quantum maps for a kicked oscillator immersed in a heat bath are obtained in both cases. These maps are studied in the classical and semiclassical limits, and it is shown that they coincide for low kicking frequency compared to the damping rate.

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I. INTRODUCTION

The macroscopic dynamics of quantal many-body systems presents a variety of realizations where a small set of collective coordinates and momenta are stochastic variables. These situations typically appear in quantum optics [1–4] and condensed-matter physics, [5, 6] and most usually, the source of external noise on the random variable is the dissipative coupling to the remaining microscopic degrees of freedom.

The formalism of quantal master equations, operating on a basic configuration consisting of a harmonic mode immersed in a heat reservoir, has proven to be a valuable tool in setting a framework for the dissipative-diffusive dynamics. In this context, most advances in the field have been provided by laser physics, bringing, as a consequence, the fact that the most accepted model for the heat bath consists of a gas of harmonic oscillators [3, 7–12]. More recently, applications to nuclear physics have been put forward [13–16] that make room for the incorporation of fermionic heat baths into the picture. An investigation of the spectral properties of the master equation in the presence of additive bosonic plus fermionic reservoirs has also been carried [17].

In view of the many important advances in the theory and applications of classical stochastic processes [18, 19], the use of semiclassical representations of quantal motion may offer several advantages, in particular, that of examining the transition to the classical regime. More recently, the Wigner transformation of the density operator and of the master equation has provided a basis for the construction of a quantum map [20], in the case where the macroscopic system is subjected to a periodic sequence of kicks in addition to the dissipative and stochastic coupling. Such a quantum map gives rise to a line of research inserted in the field of quantum chaos [20, 21] and, in particular, the relation to the Hénon map

[20] and the properties of the Lyapunov exponents [21] have been examined.

The previous work quoted here concerns a specific type of linear coupling, namely, the rotating-wave approximation (RWA), which contains only elastic vertices when the heat bath is an oscillator gas. This structure contrasts with the “fully coupled (FC) oscillator model” [9], where only the harmonic coordinate and not the momentum is affected by the interaction with the microscopic degrees of freedom. One may, however, notice that the Kramers equation of motion for the probability density in oscillator phase space $\{(Q, P)\}$ [18, 19] appears as the classical limit of the FC master equation. It is then of interest to analyze with some care the differences and similarities between the RWA and the FC models in the quantal master equations, the semiclassical representations, and the quantum maps for the kicked system.

For this reason, in Sec. II we present the derivation of the generalized master equation (GME) for the FC model and, from this equation, we obtain that corresponding to the RWA. In Sec. III, the corresponding semiclassical representations are examined and it is shown that the evolution of the FC Wigner distribution becomes asymptotically identical to the evolution of the RWA one. Following the procedure outlined by Graham and Tél for the RWA case, the FC quantum map is derived in Sec. IV, and in Sec. V, it is shown that depending on the frequency of the perturbation, the RWA description may not appear as the asymptotic limit of the FC one. The summary is presented in Sec. VI.

II. THE ASYMPTOTIC LIMIT OF THE GME

We consider a subsystem S coupled to a heat bath B with total Hamiltonian

$$H = H_S + H_B + H_{SB} \quad (1)$$

for the combined system. We will assume

$$H_{SB} = \lambda SB, \quad (2)$$

where λ scales the interaction strength and S, B are Hermitic operators on Hilbert spaces of S and B , respectively. The properties of the interacting subsystems S and B can be computed from the knowledge of either reduced density,

$$\begin{aligned} \rho_S &= \text{Tr}_B \rho, \\ \rho_B &= \text{Tr}_S \rho, \end{aligned} \quad (3)$$

where ρ is the total density operator and Tr_Q indicates tracing with respect to the quantum numbers of subsystem Q . The well-known projection techniques [22–24] yield the generalized master equation for ρ_S [3, 7, 25]. If we assume that no correlation between system and heat reservoir exists at $t = 0$, and that the reservoir is initially in thermal equilibrium, i.e.,

$$\rho_B = \rho_B^{\text{eq}} = \frac{e^{-\beta H_B}}{\text{Tr}(e^{-\beta H_B})}, \quad (4)$$

where β is the inverse temperature, the master equation for the coupling (2) takes the form [26]

$$\begin{aligned} \dot{\rho}_S(t) &= -\frac{i}{\hbar} [H_S, \rho_S(t)] - \frac{\lambda^2}{\hbar^2} \int_0^t d\tau [S, e^{-iH_S\tau/\hbar} [S, \rho_S(t-\tau)] e^{iH_S\tau/\hbar}] \text{Re}\{\Phi_B(\tau)\} \\ &\quad - \frac{i\lambda^2}{\hbar^2} \int_0^t d\tau [S, e^{-iH_S\tau/\hbar} [S, \rho_S(t-\tau)]_+ e^{iH_S\tau/\hbar}] \text{Im}\{\Phi_B(\tau)\}. \end{aligned} \quad (5)$$

In Eq. (5) the symbols $[,]$ and $[,]_+$ respectively, denote the commutator and anticommutator, while $\Phi_B(t)$ is the correlation function of the heat bath operators,

$$\phi_B(t) = \text{Tr}_B \{B(t) B \rho_B^{\text{eq}}\}, \quad (6)$$

where $B(t)$ is computed in the noninteracting scheme for the bath.

As stated in the Introduction, in the present study we will restrict ourselves to a set of specifications that allows us to work the details out in this general frame. First, the system S is chosen as a harmonic operator with frequency ω_0 and Hamiltonian

$$H_S = \frac{P^2}{2m} + \frac{m\omega_0^2}{2} Q^2. \quad (7)$$

Second, we assume that the coupling Hamiltonian takes the form

$$H_{SB}^{(\text{FC})} = \lambda QB, \quad (8)$$

where the operator B contains infinite summations of functions of all particles in the environment. In contrast to the original FC model [9] where the oscillator is coupled to an independent-oscillator reservoir, in this work no reference to an explicit model for the heat bath is made.

Finally, we will carry out our calculations in the Markovian limit [27]. In such a case, Eq. (5) admits a form amenable to time integration, actually,

$$\begin{aligned} \dot{\rho}_S(t) &+ \frac{i}{\hbar} [H_S, \rho_S(t)] \\ &= -\frac{\lambda^2}{\hbar^2} \int_0^\infty d\tau [S, [S(-\tau), \rho_S(t)]] \text{Re}\{\Phi_B(\tau)\} \\ &\quad - \frac{i\lambda^2}{\hbar^2} \int_0^\infty d\tau [S, [S(-\tau), \rho_S(t)]_+] \text{Im}\{\Phi_B(\tau)\}, \end{aligned} \quad (9)$$

where $S(-\tau) = e^{-iH_S\tau/\hbar} S e^{iH_S\tau/\hbar}$.

With this prescription, we obtain the GME

$$\begin{aligned} \dot{\rho}_S(t) &= -\frac{i}{\hbar} [H_S, \rho_S(t)] + \frac{i}{\hbar} \frac{m\omega_r^2}{2} [Q, [Q, \rho_S(t)]_+] \\ &\quad - \frac{i}{\hbar} \frac{\nu}{2m} [Q, [P, \rho_S(t)]_+] - \frac{C}{\hbar^2} [Q, [Q, \rho_S(t)]] \\ &\quad - \frac{\delta}{\hbar^2} [Q, [P, \rho_S(t)]], \end{aligned} \quad (10)$$

where we introduced the quantities ν , ω_r , C , and δ by the definitions

$$\nu = -\frac{2\lambda^2}{\hbar\omega_0} \int_0^\infty d\tau \sin(\omega_0\tau) \text{Im}\{\Phi_B(\tau)\}, \quad (11)$$

$$\omega_r^2 = \frac{2\lambda^2}{\hbar m} \int_0^\infty d\tau \cos(\omega_0\tau) \text{Im}\{\Phi_B(\tau)\}, \quad (12)$$

$$C = \lambda^2 \int_0^\infty d\tau \cos(\omega_0\tau) \text{Re}\{\Phi_B(\tau)\}, \quad (13)$$

$$\delta = -\frac{\lambda^2}{m\omega_0} \int_0^\infty d\tau \sin(\omega_0\tau) \text{Re}\{\Phi_B(\tau)\}. \quad (14)$$

We obtain an interpretation of these coefficients if we consider the evolution equations of the mean values for the first and second moments. Using that $\langle A \rangle_t = \text{Tr}_S \{A \rho_S(t)\}$ for an operator A belonging to the oscillator space, from Eq. (10) we get

$$\langle \dot{Q} \rangle_t = \frac{1}{m} \langle P \rangle_t, \quad (15)$$

$$\langle \dot{P} \rangle_t = -m\Omega^2 \langle Q \rangle_t - \frac{\nu}{m} \langle P \rangle_t, \quad (16)$$

and

$$\langle \dot{Q}^2 \rangle_t = \frac{2}{m} \langle \{QP\} \rangle_t, \quad (17)$$

$$\langle \dot{P}^2 \rangle_t = -2m\Omega^2 \langle \{QP\} \rangle_t - \frac{2\nu}{m} \langle P^2 \rangle_t + 2C, \quad (18)$$

$$\langle \{\dot{Q}P\} \rangle_t = \frac{\langle P^2 \rangle_t}{m} - m\Omega^2 \langle Q^2 \rangle_t - \frac{\nu}{m} \langle \{QP\} \rangle_t - \delta, \quad (19)$$

where $\Omega^2 = \omega_0^2 - \omega_r^2$.

Equations (15)–(19) are analogous to those obtained

in the classical case for a damped oscillator of frequency Ω , except for the presence of δ in the evolution equation for the mean value of the symmetrized product of Q and P . Therefore we can identify ω_r as a shift of the natural frequency ω_0 [8], while C and ν are the diffusion and friction coefficients, respectively. These two are related by the quantum fluctuation-dissipation relation

$$C = \frac{\hbar\omega_0}{2} \coth\left(\frac{\beta\hbar\omega_0}{2}\right) \nu, \quad (20)$$

from which

$$\begin{aligned} C + \frac{\hbar\omega_0}{2} \nu &= \hbar\omega_0 \nu [n(\omega_0) + 1], \\ C - \frac{\hbar\omega_0}{2} \nu &= \hbar\omega_0 \nu n(\omega_0), \end{aligned} \quad (21)$$

$n(\omega_0)$ being the average occupation number for oscillator quanta with frequency ω_0 at temperature $T = \beta^{-1}$. With the aid of Eq. (21), one may write the GME (10) in terms of the creation and annihilation operators,

$$\Gamma = \sqrt{\frac{m\omega_0}{2\hbar}} Q + \frac{i}{\sqrt{2m\hbar\omega_0}} P, \quad (22)$$

$$\Gamma^\dagger = \sqrt{\frac{m\omega_0}{2\hbar}} Q - \frac{i}{\sqrt{2m\hbar\omega_0}} P.$$

In this case, we get

$$\begin{aligned} \dot{\rho}_S(t) &= -i \left(\omega_0 - \frac{\omega_r^2}{2\omega_0} \right) [\Gamma^\dagger \Gamma, \rho_S] - \frac{\nu}{2m} n(\omega_0) (\Gamma \Gamma^\dagger \rho_S - 2\Gamma^\dagger \rho_S \Gamma + \rho_S \Gamma \Gamma^\dagger) - \frac{\nu}{2m} [n(\omega_0) + 1] (\Gamma^\dagger \Gamma \rho_S - 2\Gamma \rho_S \Gamma^\dagger + \rho_S \Gamma^\dagger \Gamma) \\ &\quad + \frac{\nu}{2m} n(\omega_0) ([\Gamma, \rho_S] \Gamma - \Gamma^\dagger [\Gamma^\dagger, \rho_S]) + \frac{\nu}{2m} [n(\omega_0) + 1] ([\Gamma^\dagger, \rho_S] \Gamma^\dagger - \Gamma [\Gamma, \rho_S]) \\ &\quad + \frac{i}{2\hbar} \left(\delta + \hbar \frac{\omega_r^2}{2\omega_0} \right) (\Gamma [\Gamma, \rho_S] + [\Gamma^\dagger, \rho_S] \Gamma^\dagger) - \frac{i}{2\hbar} \left(\delta - \hbar \frac{\omega_r^2}{2\omega_0} \right) (\Gamma^\dagger [\Gamma^\dagger, \rho_S] + [\Gamma, \rho_S] \Gamma). \end{aligned} \quad (23)$$

The RWA consists in ignoring those terms containing two boson operators of the same kind, i.e., the four last terms of (23). In this case, writing the result in the form of Eq. (10), one gets [28]

$$\begin{aligned} \dot{\rho}_S(t) &= -\frac{i}{\hbar} \left(1 - \frac{\omega_r^2}{2\omega_0^2} \right) [H_S, \rho_S(t)] \\ &\quad - \frac{i}{\hbar} \frac{\nu}{4m} \{ [Q, [P, \rho_S(t)]_+] - [P, [Q, \rho_S(t)]_+] \} \\ &\quad - \frac{C}{2\hbar^2} \{ [Q, [Q, \rho_S(t)]] + \frac{1}{(m\omega_0)^2} [P, [P, \rho_S(t)]] \}, \end{aligned} \quad (24)$$

Equation (24) thus appears as the long-time limit of (10) whenever the off-diagonal matrix elements of the density operator (in the Γ^\dagger, Γ representation) decay at a faster rate than the populations $\rho_n = \langle n | \rho_S | n \rangle$. This behavior can be expected in the weak-coupling limit [29] and is consistent with the adopted Markovian approximation. Consequently, hereafter we will regard the RWA master equation (24) as the asymptotic regime of the FC one (10), the latter describing the true evolution for short times, compared with the overall relaxation scale.

III. PHASE SPACE REPRESENTATION OF THE GME

In order to map the master equations for the harmonic oscillator in both the FC and RWA, we select the Wigner representation [30]. Introducing the Wigner transform of the density operator [31],

$$\rho_W(Q, P) = \int \frac{dq}{2\pi\hbar} e^{-iPq/\hbar} \left\langle Q + \frac{q}{2} \left| \rho_S \right| Q - \frac{q}{2} \right\rangle \quad (25)$$

and employing the well-known rule of operator products [31],

$$\begin{aligned} (\hat{A}\hat{B})_W(Q, P) &= A_W(Q, P) \exp \left[\frac{\hbar}{2i} \left(\frac{\overleftarrow{\partial}}{\partial P} \frac{\overrightarrow{\partial}}{\partial Q} - \frac{\overleftarrow{\partial}}{\partial Q} \frac{\overrightarrow{\partial}}{\partial P} \right) \right] \\ &\quad \times B_W(Q, P), \end{aligned} \quad (26)$$

some straightforward algebra easily leads to the semiclassical equations of motion,

$$\begin{aligned} \frac{\partial}{\partial t} \rho_W^{(\text{FC})}(Q, P, t) &= \left\{ -\frac{\partial}{\partial Q} \frac{P}{m} + \frac{\partial}{\partial P} \left(m\Omega^2 Q + \frac{\nu}{m} P \right) \right. \\ &\quad \left. - \delta \frac{\partial^2}{\partial Q \partial P} + C \frac{\partial^2}{\partial P^2} \right\} \rho_W^{(\text{FC})}(Q, P, t), \end{aligned} \quad (27)$$

and

$$\begin{aligned} \frac{\partial}{\partial t} \rho_W^{(\text{RWA})}(Q, P, t) &= \left\{ -\frac{\partial}{\partial Q} \left[\left(1 - \frac{\omega_r^2}{2\omega_0^2} \right) \frac{P}{m} - \frac{\nu}{2m} Q \right] \right. \\ &\quad \left. + \frac{\partial}{\partial P} \left[m \left(\omega_0^2 - \frac{\omega_r^2}{2} \right) Q + \frac{\nu}{2m} P \right] \right. \\ &\quad \left. + \frac{\partial^2}{\partial Q^2} \frac{C}{2m^2\omega_0^2} + \frac{\partial^2}{\partial P^2} C \right\} \\ &\quad \times \rho_W^{(\text{RWA})}(Q, P, t). \end{aligned} \quad (28)$$

These equations are exact representations of the FC and RWA master equations (10) and (24) in $\{(Q, P)\}$ phase space. It is worthwhile noticing that, in the classical limit, δ approach zero while

$$\lim_{\beta\hbar\omega_0 \rightarrow 0} C = \nu T \quad (29)$$

In such a case, Eq. (27) is precisely Kramers's equation [19] describing the motion of the probability density of a classical oscillator of frequency Ω undergoing Brownian motion. We are then led to the conclusion that it is the FC, rather than the RWA, that is the quantal source of the appropriate classical limit. On the other hand, some other important differences between Eqs. (27) and (28) may be visualized from the examination of the respective equilibrium solutions. In either case, one easily finds the equilibrium distribution looking for a general form,

$$\rho_W^{\text{eq}} \propto \exp\left(-\frac{Q^2}{2\sigma_{QQ}^2} - \frac{P^2}{2\sigma_{PP}^2}\right), \quad (30)$$

which, after replacement, gives rise to equilibrium dispersions, in the FC

$$\begin{aligned} \sigma_{QQ}^2 &= \frac{C - \nu\delta}{m\nu\Omega^2}, \\ \sigma_{PP}^2 &= m\frac{C}{\nu}, \end{aligned} \quad (31)$$

and, in the RWA

$$\begin{aligned} \sigma_{QQ}^2 &= \frac{1}{m\omega_0^2} \frac{C}{\nu}, \\ \sigma_{PP}^2 &= m\frac{C}{\nu}. \end{aligned} \quad (32)$$

The semiclassical representation of the RWA master equation has been investigated in Ref. [32], where it has been shown by explicit calculations that in actionlike variables (H, ϕ) , Eq. (28) takes the form

$$\begin{aligned} \frac{\partial}{\partial t} \rho_W^{(\text{RWA})}(H, t) &= \left\{ \frac{\partial}{\partial H} \left(\frac{\nu}{m} H - \frac{C}{m} \right) \right. \\ &\quad \left. + \frac{\partial^2}{\partial H^2} \frac{C}{m} \right\} \rho_W^{(\text{RWA})}(H, t), \end{aligned} \quad (33)$$

in the current notation. It corresponds to the asymptotic regime described by the diagonal part of the FC master equation (see the discussion at the end of the previous section), where only occupation probabilities of oscillator eigenstates evolve in time. By contrast, the action-angle version of the FC semiclassical motion (27) reads

$$\begin{aligned} \frac{\partial}{\partial t} \rho_W^{(\text{FC})}(H, \phi, t) &= \left\{ -\frac{\partial}{\partial H} \left(\frac{C}{m} - 2\frac{\nu}{m} H \sin^2 \phi + \frac{\omega_r^2}{\omega_0} H \sin 2\phi \right) \right. \\ &\quad - \frac{\partial}{\partial \phi} \left[-\omega_0 \left(1 - \frac{\omega_r^2}{\omega_0^2} \cos^2 \phi \right) - \frac{\nu}{2m} \left(1 - \frac{C}{\nu H} \right) \sin 2\phi + \frac{\delta\omega_0}{2H} \cos 2\phi \right] \\ &\quad + \frac{\partial^2}{\partial \phi \partial H} \left(\frac{C}{2m} \sin 2\phi - \frac{\delta\omega_0}{2} \cos 2\phi \right) + \frac{\partial^2}{\partial \phi^2} \left(\frac{C}{2mH} \cos^2 \phi + \frac{\delta\omega_0}{2H} \cos 2\phi \right) \\ &\quad \left. + \frac{\partial^2}{\partial H^2} \left(2\frac{C}{m} H \sin^2 \phi \right) \right\} \rho_W^{(\text{FC})}(H, \phi, t). \end{aligned} \quad (34)$$

A comparison between Eqs. (34) and (33) brings into evidence that (33) appears as the angular average of (34), provided that $\rho_W^{(\text{FC})}$ does not depend on the angle $\phi = \tan^{-1}(P/m\omega_0 Q)$; in other words, if the Wigner distribution only represents a probability density on the energy variable. This consideration leads us again to the asymptotic regime of FC; however, the apparent inconsistent results for the dispersions displayed in Eqs. (31) and (32) remain unexplained.

To advance one more step in the resolution of this discrepancy while shedding light upon the conditions that specify the asymptotic regime of the FC, we shall employ a method similar to the one introduced by Stratonovich [33] to investigate a nonlinear oscillator with additive noise in the low friction limit. The idea is to exploit the presence of two well-separated time scales, on the basis that if the damping rate is low, the average energy

envelope undergoes small variations over one period of the average displacement. The method is analogous to adiabatic elimination of fast variables [34] in a scheme where energy is a slow variable while position and momentum are fast ones; in terms of the density operator, this philosophy corresponds to considering a situation where only diagonal matrix elements in the Hamiltonian basis $|n\rangle$ evolve, since the off-diagonal ones of the type $\rho_{n, n\pm 1}$ needed to set the average position and momentum have vanished in a shorter time scale.

The procedure is as follows. We consider Eq. (27) and switch variables (Q, P) to (Q, H) . Now, we assume that $\rho_W(Q, H, t)$ can be written as a probability density for conditional events,

$$\rho_W(Q, H, t) = W(H, t)W'(Q, t/H), \quad (35)$$

where $W(H, t)$ is the probability density for the oscilla-

tor to have energy H at time t , and $W(Q, t/H)$ is the conditional probability density for the coordinate to be Q if the energy is H at the given time.

It is then natural to assume that the transition probability $W(Q, t/H)$ is proportional to the time that the oscillator spends at the position Q while having energy H ; in other words, while having velocity $P(Q, H) = \sqrt{2mH - m^2\omega_0^2 Q^2}$. We then write (35) as

$$\rho_W(Q, H, t) = \frac{m\omega_0}{\pi P(Q, H)} W(H, t), \quad (36)$$

where the normalization factor has been chosen to guarantee that $W(H, t)$ is the marginal distribution,

$$W(H, t) = \int_{R_H} dQ \rho_W(Q, H, t) \quad (37)$$

over the region $R_H = \{Q | \frac{1}{2}m\omega_0^2 Q^2 \leq H\}$. It should be noted that (36) is exact at $t \rightarrow \infty$. Carrying out the change of variables in Eq. (27) and replacing in (37), one finds, after suppressing vanishing integrals,

$$\begin{aligned} \frac{\partial}{\partial t} W(H, t) &= \frac{\omega_0}{\pi} \left\{ \frac{\partial}{\partial H} \int_{R_H} dQ \left(\frac{\nu}{m} P(Q, H) - \frac{C}{P(Q, H)} \right) \right. \\ &\quad \left. + \frac{\partial^2}{\partial H^2} \int_{R_H} dQ \frac{C}{m} P(Q, H) \right\} W(H, t). \end{aligned} \quad (38)$$

Since the value of the action over one orbit $H = \text{const}$ is

$$S(H) = \int_{R_H} dQ P = \frac{\pi}{\omega_0} H, \quad (39)$$

one readily finds that Eq. (38) is identical to (33).

The conclusion of this calculation is that the semiclassical representation of the RWA master equation is the long-time limit of the corresponding FC description in

the low friction limit. The latter condition is in turn related to the weak-coupling regime, where one is able to prove the equivalent relationship between the quantal RWA and FC evolutions. In such a case, both the coefficient δ and the frequency shift are negligible; thus the FC and RWA dispersions in Eqs. (32) and (31) can be regarded as essentially identical. It then becomes clear that asymptotic harmonic quantal Brownian motion can be mapped onto phase space without reaching any inconsistency, while the classical Kramers equation appears as the semiclassical version of the FC and governs the evolution of the probability over the whole time axis.

IV. THE QUANTAL MAP FOR A KICKED SYSTEM

We will now apply the concepts discussed in the previous sections to a periodically forced oscillator immersed in a heat reservoir. Such a system can be modeled by the Hamiltonian,

$$H' = H - \sum_{n=-\infty}^{\infty} \delta(t - n\tau) g(Q, P), \quad (40)$$

where H is given by (1) and $g(Q, P)$ gives the strength of the kicks whose period is τ . For simplicity we will consider this intensity to be only a function of the position Q .

Graham and Tél [20] have derived a quantum map for the kicked oscillator when the coupling mechanism corresponds to the RWA. The procedure can be summarized as follows. As indicated by several authors [35, 36] the dynamics of the pulsating system undergoes two regimes: (i) between kicks, where the motion of the density matrix is described by the master equation, and (ii) around the kicks ($t \sim n\tau$), where the periodic term in (40) dominates overall propagation. The change in the density matrix due to the n th kick can be expressed as

$$\rho(n\tau^+) = \exp\left(\frac{i}{\hbar} g(Q)\right) \rho(n\tau^-) \exp\left(-\frac{i}{\hbar} g(Q)\right). \quad (41)$$

From the definition (25) of the Wigner function, it follows that

$$\rho_W(Q, P, n\tau^+) = \int \frac{dq}{2\pi\hbar} \exp\left(-\frac{i}{\hbar} Pq\right) \exp\left\{\frac{i}{\hbar} \left[g\left(Q + \frac{q}{2}\right) - g\left(Q - \frac{q}{2}\right)\right]\right\} \left\langle Q + \frac{q}{2} \left| \rho(n\tau^-) \right| Q - \frac{q}{2} \right\rangle \quad (42)$$

which, using the inverse transformation, can be set as

$$\rho_W(Q, P, n\tau^+) = \iint \frac{dq dp}{2\pi\hbar} \exp\left(-\frac{i}{\hbar} Pq\right) \exp\left\{\frac{i}{\hbar} \left[g\left(Q + \frac{q}{2}\right) - g\left(Q - \frac{q}{2}\right)\right]\right\} \rho_W(Q, P, n\tau^-) \quad (43)$$

In phase space, the evolution of the oscillator between kicks is given by Eq. (28). An exact solution for the latter can be written in terms of the generating function,

$$\phi^{(\text{RWA})}(\psi, \eta, t) = \iint dQ dP \exp\{i(\psi Q + \eta P)\} \rho_W^{(\text{RWA})}(Q, P, t). \quad (44)$$

If one performs the transformation (44) on Eq. (28), a first-order partial derivative equation for $\phi(\psi, \eta, t)$ is encoun-

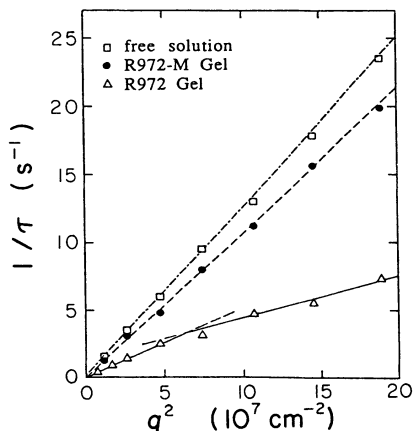


FIG. 4. Plot of the decay rate $1/\tau$ of the transient diffraction from the grating of the excited dyes attached to a polystyrene (MW of 1.3×10^6) vs the squared wave vector q^2 in three different systems: (i) unbounded solution (\square), (ii) silica gel R972-M (\bullet), and (iii) silica gel R972 (\triangle). Proportionality exists in both the unbounded solution and the R972-M gel, a relation indicating Brownian motion, whereas an obvious curvature is observed in the R972 gel. The slope of $1/\tau$ vs q^2 defines the diffusion coefficient for systems (i) and (ii). The quotient $1/\tau$ divided by q^2 at $q^2 = 4.74 \times 10^7 \text{ cm}^{-2}$ is used as the apparent diffusion coefficient D_a to describe the porous medium R972.

angle measurement at $q^2 = 4.74 \times 10^7 \text{ cm}^{-2}$ (corresponding to a prism spacing of 25 cm), where $D_a = 1/(\tau q^2)$.

Polymer diffusion within a silica gel or a silica suspension is hindered relative to that in the unbounded solution. The ratio of the diffusion coefficient inside the porous medium R972-M to that in the unbounded solution D/D_0 (a ratio often referred to as the hindrance factor) is plotted in Fig. 5 as a function of silica volume fraction Φ . The general trend is that the value of D/D_0 decreases with increasing Φ , thus indicating a stronger hin-

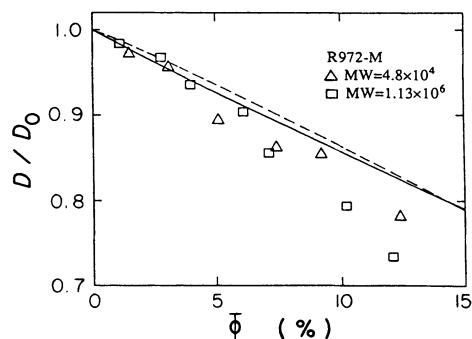


FIG. 5. Hindrance factor D/D_0 vs silica volume fraction Φ for two polystyrene samples (MW of 4.8×10^4 and 1.13×10^6) in R972-M solutions and gels. The gelation threshold is approximately $\Phi = 6\%$. The weak hindrance is attributed to geometric obstruction. The solid curve represents the theory of Neale and Nader [27] [Eq. (3)] for diffusion in a homogeneous, isotropic swarm of spheres. The dashed curve corresponds to the bound predicted by Prager [28] [Eq. (4)] for a homogeneous, isotropic suspension of solid particles of arbitrary shape.

drance. In the low silica concentration ($\Phi < 6\%$) regime, the silica suspension will not gel, and the diffusion coefficient was measured before the separation of the dilute silica suspension into two layers (as a result of precipitation). In the higher silica concentration ($\Phi > 6\%$) regime, the diffusion coefficients were measured after the gels had formed.

Two labeled polystyrene samples with different molecular weights (MW of 4.8×10^4 and 1.13×10^6) were used in this study. We note that there is little difference (within experimental error) between the hindrance values for these two polymer sizes. This observation implies that the polymer molecules are still much smaller than the pore dimensions. In other words, the polystyrene molecules of both sizes can still be approximated as point particles. The use of even higher-molecular-weight polymers, although desirable, is limited by our ability to label larger molecules.

Labeled polystyrene molecules diffuse much faster in the porous medium R972-M than in the R972 porous medium (see Fig. 6), where adsorption severely reduced the freedom of the polymer. The weak hindrance in R972-M is relatively more dependent on Φ than is the diffusion behavior in R972 (see Fig. 6). These facts suggest that the effect of adsorption is negligible in R972-M, and that the hindrance can be attributed, for the most part, to geometric obstruction and hydrodynamic interaction with the silica surfaces. There exist many theories that take these two factors into account and that predict the relation between the hindrance factor D/D_0 and the porosity for different porous systems. Our experimental results are compared with the hydrodynamic theory developed by Neale and Nader [27] for a much simpler model system—that of a homogeneous swarm of spherical particles of arbitrary size distribution. Based on the nonrigorous assumption that each particle experiences the remainder as a uniform fluid, this model predicts that

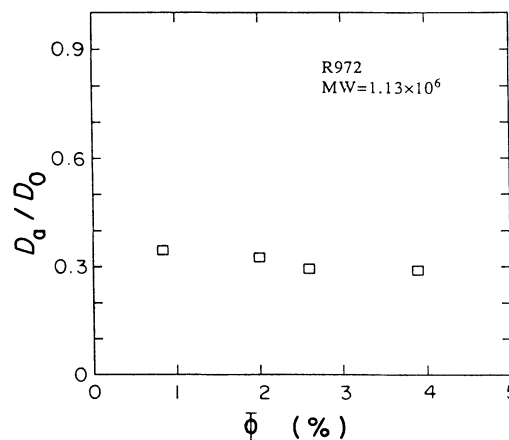


FIG. 6. Hindrance factor D_a/D_0 vs silica volume fraction Φ for polystyrene (MW of 1.13×10^6) in R972 solutions and gels. The gelatin threshold is approximately $\Phi = 2.2\%$. The polymer diffusion is strongly hindered even at low Φ . This hindrance is only slightly dependent on Φ .

$$\frac{D}{D_0} = \frac{2(1-\Phi)}{2+\Phi} \quad (3)$$

(Note that in Neale and Nader's original notation, Φ was used to denote porosity, which we denote as $1-\Phi$.) Equation (3), independent of the size distribution of the spheres, is in satisfactory agreement with the experimental data for diffusion in a wide range of porous media throughout the whole porosity range. The solid line in Fig. 5 represents Eq. (3). Our results in the low- Φ regime are reasonably consistent (within experimental error) with the Neale-Nader theory [Eq. (3)]. This correlation can be explained by the fact that the silica suspensions can be regarded approximately as a collection of basic spherical units, even though these units had aggregated into ramified clusters. At low solid-volume fraction Φ (or high porosity), the trend of hindered diffusion in a fumed silica gel is similar to that in an unconsolidated homogeneous swarm of spherical particles.

In the gelled systems, the formation of three-dimensional networks further deviates from the assumption of the Neale-Nader model that requires different particles in the suspension to be independent of one another in the sense that their regions of hydrodynamic influence must not overlap. This deviation is greater at higher silica concentration. We observe that at higher Φ (or lower porosity), the hindered diffusion tends to be slower than that predicted by the Neale-Nader theory. We emphasize here that the Neale-Nader model does not totally parallel our experimental system. Therefore, this comparison (as well as another comparison to follow) is thus intended only to put this work in a more general context of studies of hindered transport in random porous media.

Our results are also compared with a theory developed by Prager [28] for a homogeneous and isotropic suspension of solid particles of arbitrary shape. In this treatment, the main assumptions in the Neale-Nader model were relaxed, and the principle of minimum entropy production was applied to obtain bounds of the hindered diffusion rate:

$$\frac{D}{D_0} < (1-\Phi) \left[1 - \frac{\Phi}{3} \right] \quad (4)$$

(Again, Φ is the silica volume fraction rather than porosity, as denoted by Prager.) The dashed curve in Fig. 5 corresponds to Prager's theory [Eq. (4)]. Our experimental results obviously satisfy the inequality relation predicted by Prager.

The ratio of diffusion coefficient D_a/D_0 for labeled polystyrene (MW of 1.13×10^6) in the porous media of silica R972 is plotted against silica volume fraction Φ in Fig. 6, where D_a is as defined in Sec. II. All data in this figure were taken after the diffusion coefficient had stabilized; as will be demonstrated, the diffusion rate decreases soon after mixing the silica suspension. We found that the value of D_a/D_0 is only slightly dependent on the silica fraction Φ . This observation is explained by the fact that adsorption plays a dominant role in slowing down polymer diffusion and that even at low concentration the silica surface area is greater than that which would be

covered by the adsorbed polymer molecules. Yet the measured diffusion coefficient in the R972 gel still decreases by a small amount as Φ increases. There are three possibilities to account for this small decrease: (i) the increased ratio of surface-to-pore volume results in a higher probability of adsorption; (ii) geometric obstruction, such as tortuosity, is increased; and (iii) more silica particles cause more static scattering and stray light, which produce a poorer signal-to-noise ratio and a higher baseline value, thus introducing greater error into the data analysis. We also found that, at the two lower silica concentrations, the silica suspension will not gel and the hindrance in these two suspensions is similar to that in the gels.

Although the labeling ratio or the density of the dye segments does not affect the diffusion rate in the R972-M porous medium, it does have an effect on the diffusion coefficient for the dye-labeled polystyrene in the R972 porous medium. For polymer samples with the same molecular weight, a higher labeling ratio usually results in slower diffusion. Thus, comparison between different polymer sizes is not made, as there is no quantitative control over the labeling ratio in the labeling reactions. As an approximate generalization of this observation, for most molecular weights and labeling ratios studied, the value of D_a/D_0 is between 0.2 and 0.4 in a Φ range of about 1–5%.

The silica gels in this work can be viewed as physical gels that are reversible in nature, because there is no chemical bonding involved in gelation. We monitored the development of hindrance during the formation of silica gels. Initially, the mixture of silica and the polymer solution was vigorously shaken on a model Vortex-Genie test tube mixer to destroy the interconnection of the silica particles. FRS measurements were then conducted to obtain the diffusion coefficient at different times during the gelation process. We observed little difference (within experimental error) between a "fresh" gel and a gel formed from a "destroyed" one.

The diffusion coefficient for polystyrene (MW of 1.13×10^6) inside a R972-M gel ($\Phi = 7.1\%$) as a function of gelation time is shown in Fig. 7. There is a slight

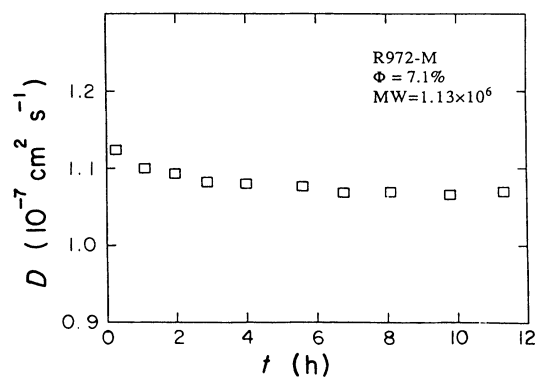


FIG. 7. Diffusion coefficient D vs time during the gelation process for an R972-M gel ($\Phi = 7.1\%$). The time for stabilization of the diffusion coefficient (approximately 5 h) is comparable to that needed for the silica suspension to gel.

This expression has been derived by Graham and Tél, who call the attention upon the fact that the only effect of the kick appears in the classical Langevin map $(Q_n^{(RWA)}, P_n^{(RWA)})$ displayed in Eqs. (54), i.e., the impulses just provoke classical noise. The same concept applies to the classical limit of the FC quantum map; indeed, if one integrates Eq. (60), one finds

$$K_{\text{class}}^{(\text{FC})}(Q, P, Q_{n-1}, P_{n-1}) = \frac{1}{2\pi\sigma(\tau)} \exp\left(-\frac{1}{2\sigma^2(\tau)}[(Q - Q_n^{(\text{FC})})^2\sigma_{PP}^2(\tau) + (P - P_n^{(\text{FC})})^2\sigma_{QQ}^2(\tau) - 2\sigma_{QP}^2(\tau)(Q - Q_n^{(\text{FC})})(P - P_n^{(\text{FC})})]\right) \quad (65)$$

with

$$\sigma^2(\tau) = \sigma_{QQ}^2(\tau)\sigma_{PP}^2(\tau) - \sigma_{QP}^4(\tau). \quad (66)$$

The classical Langevin map $(Q_n^{(\text{FC})}, P_n^{(\text{FC})})$ of Eqs. (61) now corresponds to harmonic Brownian motion. Several points may be stressed in light of expressions (64) and (65). First, one notices that in the absence of any periodic perturbation, replacing the maps $(Q_n^{(\alpha)}, P_n^{(\alpha)})$ in the arguments of the Gaussian functions by the continuous functions (50) or (56) leads to the kernels of the Wigner functions $\rho_W^{(\alpha)}(Q, P)$ under the free propagator indicated by (27) and (28), respectively. Since the asymptotic dispersions $\sigma_{QQ}^2(t)$, $\sigma_{PP}^2(t)$ and $\sigma_{QP}^2(t)$ are the equilibrium values σ_{QQ}^2 , σ_{PP}^2 , and 0, it is clear that (64) is the long-time version of (65), as expected according to the discussion in Sec. III. A similar relationship between both maps may be encountered when $\frac{\nu\tau}{2m} \gg 1$ ($E \rightarrow 1$). In such a

case, the FC and RWA kernels coincide with the dispersions set at their asymptotic values. This is a situation characterized by a low kicking frequency, as compared to the damping rate; consequently, the propagator between kicks takes long enough time to reach its free asymptotic regime before receiving any impulsion. In the opposite limit, $\frac{\nu\tau}{2m} \ll 1$ ($E \rightarrow 0$), the damping strength is negligible and $\rho_W^{(\text{RWA})}(Q, P)$ is unable to freely drift or diffuse; the perturbation thus freezes this Wigner distribution at the starting one, $\rho_W^{(\text{RWA})}(Q_n^{(\text{RWA})}, P_n^{(\text{RWA})})$. The FC kernel, however, remains finite and displays a nonvanishing Q - P correlation. Throughout the full range of intermediate ratios $\frac{\nu\tau}{2m}$, the RWA and FC do not coincide.

It is not possible to advance this comparison beyond the classical limit, due to the complication introduced by the phase function G given in (63). The lowest-order correction proportional to \hbar^2 has been displayed in Ref. [20] for the RWA case. In the current notation, it reads

$$K^{(\text{RWA})}(Q, P, Q_{n-1}, P_{n-1}) = \frac{\sqrt{2}}{\sqrt{\pi}[\hbar^2 |g'''(Q)|]^{1/3}\sigma_{QQ}(1-E)^{1/2}} \exp\left(\frac{(Q - Q_n^{(\text{RWA})})^2}{2\sigma_{QQ}^2(1-E)}\right) \exp\left(\frac{16}{3\hbar^4} \frac{\sigma_{PP}^6(1-E)^3}{g'''(Q)^2}\right) \times \exp\left\{\frac{4\sigma_{PP}^2(1-E)}{\hbar^2 g'''(Q)}(P - P_n^{(\text{RWA})})\right\} \text{Ai}\left(\left(P - P_n^{(\text{RWA})} + \frac{2\sigma_{PP}^4(1-E)^2}{\hbar^2 g'''(Q)}\right) \frac{2}{[\hbar^2 g'''(Q)]^{1/3}}\right), \quad (67)$$

where Ai stands for the Airy functions [37].

If one computes the FC kernel under the same approximation, one finds

$$K^{(\text{FC})}(Q, P, Q_{n-1}, P_{n-1}) = \frac{\sqrt{2}}{\sqrt{\pi}[\hbar^2 |g'''(Q)|]^{1/3}\sigma_{QQ}(\tau)} \exp\left(\frac{(Q - Q_n^{(\text{FC})})^2}{2\sigma_{QQ}^2(\tau)}\right) \times \exp\left[\frac{16}{3\hbar^4} \frac{\sigma'^6(\tau)}{g'''(Q)^2} + \frac{4\sigma'^2(\tau)}{\hbar^2 g'''(Q)} \left(P - P_n^{(\text{FC})} - \frac{\sigma_{QP}^2(\tau)}{\sigma_{QQ}^2(\tau)}(Q - Q_n^{(\text{FC})})\right)\right] \times \text{Ai}\left\{\left(P - P_n^{(\text{FC})} - \frac{\sigma_{QP}^2(\tau)}{\sigma_{QQ}^2(\tau)}(Q - Q_n^{(\text{FC})}) + \frac{2\sigma'^4(\tau)}{\hbar^2 g'''(Q)}\right) \frac{2}{[\hbar^2 g'''(Q)]^{1/3}}\right\}, \quad (68)$$

with

$$\sigma'(\tau)^2 = \sigma_{PP}^2(\tau) - \frac{\sigma_{QP}^4(\tau)}{\sigma_{QQ}^2(\tau)}. \quad (69)$$

Once again, the major differences may be ascribed to the appearance of the correlation $\sigma_{QP}^2(\tau)$ and nonequilibrium dispersions $\sigma_{QQ}^2(\tau)$, $\sigma_{PP}^2(\tau)$. These modified propagators can be seen to coincide in the same situations encountered in the classical limit, as expected.

VI. SUMMARY

In this work, we have examined the relationships between two descriptions of a quantal harmonic oscillator coupled to a random dissipative reservoir, namely, the FC and RWA, in three different frames and without making any specific assumption on the nature of the heat bath.

First, we have reviewed the approach based on quantal master equations, recalling the fact that the RWA version appears as a long-time limit of the FC one, provided that the coupling is weak — an assumption that, on the other hand, enables one to restrict oneself to the Markovian regime. Second, we have established the semiclassical representation of either master equation in terms of the corresponding Wigner quasidistribution. In this case, by resorting to an adiabatic elimination procedure, it is also possible to demonstrate that the weak-coupling plus long-time regime of the FC semiclassical dynamics is the semiclassical mapping of the RWA quantal motion. This procedure makes evident the kind of information that is lost as one approaches the asymptotic evolution of the stochastic, damped variables, namely, the one associated with coordinate and momentum — a fact that, from a

different viewpoint, can be visualized as one recognizes the existence of separated quantum scales for the decay of diagonal and nondiagonal matrix elements of the density operator.

Finally, we have looked for the quantum maps in the spirit of Refs. [36, 20], arising as one adds a periodic sequence of kicks to the already existing coupling. The kernels of the FC and RWA Wigner functions can be seen to coincide for the asymptotic regime in the semiclassical limit, where the periodic impulsion becomes the source of extra noise, as put forward by Graham and Tél. However, the asymptotic equivalence may be destroyed if the kicking frequency is sufficiently high; in such a case, a clear distinction between the coupling mechanisms described in the RWA and the FC can be pointed out.

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