Phase space dynamics of overdamped quantum systems

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Abstract. – The phase space dynamics of dissipative quantum systems in strongly condensed phase is considered. Based on the exact path integral approach, it is shown that the Wigner transform of the reduced density matrix obeys a time evolution equation of Fokker-Planck type valid from high down to very low temperatures. The effect of quantum fluctuations is discussed and the accuracy of these findings is tested against exact data for a harmonic system.

Introduction. – Dynamics in strongly condensed phase can be found almost everywhere in nature, e.g. for tunnel diodes in mesoscopic physics, for macromolecules in biological and soft-matter systems, and for chemical reactions, to name but a few. Associated with this bunch of realizations is a rich phenomenology comprising prominent effects such as stochastic resonance [1], resonant activation [2], transport in ratchets [3], and adiabatic electron transfer [4] which have been explored extensively in the last decade. However, most of these studies have focused on the domain of classical physics and less is known about low-temperature quantum properties. This is mainly due to the fact that, in contrast to the Fokker-Planck equation for the classical phase space distribution [5], a simple time evolution equation for the reduced density matrix of a dissipative quantum system does not exist in general [6]. Typically, quantum fluctuations appear on a time scale $\hbar \beta (\beta = 1/k_B T)$ so that at lower temperatures the quantum stochastic process becomes strongly non-Markovian and intimately depends on the initial correlations between system and heat bath. An exact expression for the density matrix is available within the path integral formulation, but numerical evaluations are in most cases prohibitive [6].

In the low-friction limit, e.g. for quantum optical systems, progress can be made by a Born-Markov approximation leading to well-known master equations. The opposite limit of strong damping has only recently attracted considerable attention [7–10]. In particular, it was shown [8] that for strong friction and low temperatures the density matrix is essentially restricted to its diagonal part, the position probability distribution, and that this part obeys a quantum analogue of the classical Smoluchowski equation. The question we address here goes far beyond: Is there a time evolution equation in whole phase space valid from high down to low temperatures provided friction is sufficiently strong? The answer is yes, and we will show this in detail below.
Path integral formulation. – The inclusion of dissipation within quantum mechanics is well established and is based on a system + reservoir formulation \([6]\). The dynamics of the corresponding density matrix starting at \(t = 0\) from a general initial state \(W(0)\) reads
\[
W(t) = \exp[-iHt/\hbar]W(0)\exp[iHt/\hbar],
\]
where the Hamiltonian \(H = H_S + H_R + H_I\) contains a system, a reservoir (heat bath), and a system-reservoir interaction part, respectively. Dissipation arises as the effective impact of the reservoir degrees of freedom on the system dynamics within a reduced picture \(\rho(t) = \text{tr}_R\{W(t)\}\). In the standard description the Gaussian statistics of the heat bath is modeled by a quasi-continuum of harmonic oscillators bilinearly coupled with the system. In fact, this way one regains in the classical limit the generalized Langevin equation of Brownian motion.

The only non-perturbative treatment of the system-reservoir coupling is an exact elimination of the bath degrees of freedom by means of the path integral approach. For the position representation of the reduced density matrix one obtains
\[
\rho(q_f, q'_f, t) = \int dq_i dq'_{i} J(q_f, q'_f, t, q_i, q'_i) \lambda(q_i, q'_i).
\]

Here, the propagating function \(J(\cdot)\) is a threefold path integral over the system degrees of freedom only. The two real time paths \(q(s)\) and \(q'(s)\) connect in time \(t\) the initial points \(q_i\) and \(q'_i\), with the fixed end points \(q_f\) and \(q'_f\), while the imaginary time path \(q\) runs from \(q_i\) to \(q'_i\) in the interval \(h\beta\). The contribution of each path is weighted by \(\exp[i\Sigma[q, q', \bar{q})/\hbar]\) with an effective action \(\Sigma[q, q', \bar{q}]\) not specified here explicitly. Basically, it comprises the actions of the bare system in imaginary and real time, respectively, and additional interaction contributions (influence functional), non-local in time, which in the reduced picture rule the influence of the reservoir onto the system. In the limit \(t \to 0\), one has \(J(q, q', t, q_i, q'_i) \to \rho_\beta(q, q'_i)\delta(q_t - q_i)\delta(q'_t - q'_i)\) so that
\[
\rho(q_f, q'_f, 0) = \rho_\beta(q_f, q'_f) \lambda(q_i, q'_i)
\]
with the reduced equilibrium density matrix \(\rho_\beta(q, q') = \langle q|\text{tr}_R \exp[-\beta H]|q'\rangle\) and a preparation function \(\lambda\) characterizing initial deviations from thermal equilibrium. The crucial point here is that eq. (3) is a correlated initial state \([11]\), in contrast to a factorizing initial state used in ordinary Feynman-Vernon theory. In the strong-friction regime the latter one is not applicable in contrast to what has been done in \([10]\). Even in the classical limit does a product initial state not allow for a consistent derivation of the well-known Smoluchowski equation.

Overdamped quantum systems. – The stochastic dynamics of dissipative quantum systems determined by eqs. (2) and (3) is rather subtle as it strongly depends on initial correlations between system and reservoir as well as on the non-local self-interactions contained in the influence functional \([11]\). Both are governed by the damping kernel

\[
K(\theta) = \frac{\int_0^{\infty} d\omega}{\pi} \frac{\cosh[\omega(h\beta/2 - i\theta)]}{\sinh(\omega h\beta/2)},
\]

where \(\theta = s - \tau, 0 \leq s \leq t, 0 \leq \tau \leq h\beta\) and \(I(\omega)\) is the spectral density of the heat bath. Accordingly, the impact of the bath is completely specified by temperature \(T\) and \(I(\omega)\). In particular, the spectral density is related to the macroscopic damping via
\[
\gamma(s) = \frac{2}{M} \frac{\int_0^{\infty} d\omega}{\pi} \frac{I(\omega)}{\omega} \cos(\omega s).
\]
Now, we turn to the simplifications that arise in the overdamped regime. For this purpose we define a typical damping strength as

$$\gamma \equiv \hat{\gamma}(0) = \lim_{\omega \to 0} \frac{I(\omega)}{M \omega}$$

(6)

with $\hat{\gamma}(\omega)$ the Laplace transform of $\gamma(t)$. For instance, in the Ohmic case $I(\omega) = M \gamma \omega$ and also for the more realistic Drude model $I(\omega) = M \gamma \omega^2 / (\omega^2 + \omega_c^2)$ (cut-off frequency $\omega_c$), one finds $\gamma = \hat{\gamma}$. Given a typical frequency $\omega_0$ of the bare system, e.g. the ground-state frequency, by strong damping we then mean

$$\frac{\gamma^2}{\omega_0^2} \gg \hbar, \frac{1}{\omega_c}, \frac{1}{\gamma}.$$  

(7)

In other words, we assume the time scale separation well known from the classical overdamped regime [5] and extend it to the quantum range by incorporating the time scale for quantum fluctuations $\hbar \beta$. Correspondingly, we consider the dynamics equation (2) on the coarse-grained time scale $s \gg \hbar \beta, \frac{1}{\omega_c}, \frac{1}{\gamma}$ and $\sigma \gg \frac{1}{\omega_c}, \frac{1}{\gamma}$. The consequences are substantial: i) the strong friction suppresses non-diagonal elements of the reduced density matrix during the time evolution. This simply reflects the fact that a quantum system behaves more classically, and the stronger coherences are destroyed by the presence of a heat bath. ii) The real-time part $K(s)$ of the damping kernel becomes local on the coarse-grained time scale so that a time evolution equation of the form $\dot{\rho}(t) = \mathcal{L} \rho(t)$ with a time-independent operator $\mathcal{L}$ may exist.

Following the above simplifications, the path integral formulation now allows for a perturbative treatment in the strong-damping limit. To this end, it is convenient to introduce sum and difference coordinates for the imaginary time path, $\bar{\mathbf{r}} = (\bar{q} + \bar{q}')/2$ and $\bar{x} = \bar{q} - \bar{q}'$, and sum and difference paths in real time, $r(s) = [q(s) + q'(s)]/2$ and $x(s) = q(s) - q'(s)$, respectively. The idea is to evaluate the path integrals in the sense of a semiclassical approximation by assuming self-consistently that non-diagonal elements, i.e. $\bar{x}$- and $x(s)$-dependent terms, remain small during the time evolution. Hence the effective action $\Sigma[r, x, \bar{q}]$ is expanded up to second order in the $\bar{x}$ coordinate of the imaginary time path and in the excursions $x(s)$ of the real-time path integrals. Doing so, we take sufficiently smooth potentials for granted. While this procedure is applicable to a wide range of spectral bath densities, we concentrate in the sequel on the quasi-Ohmic case with a very large cut-off frequency $\omega_c \gg \gamma$. It is worthwhile to note that we do not restrict the value of $\gamma/h \beta$ meaning that our analysis covers a broad temperature range from the classical ($\gamma/h \beta \ll 1$) to the deep quantum domain ($\gamma/h \beta \gg 1$).

**Equilibrium density matrix.** – The question to what extent the thermodynamic equilibrium is affected by the strong-damping limit, particularly at lower temperatures, is a very crucial one and also serves as a basis for the more involved treatment of the dynamical case. In terms of a sum over paths from $\bar{q}$ to $\bar{q}'$ in the time interval $[0, h \beta]$ the unnormalized reduced equilibrium density matrix in position representation reads

$$\rho_\beta(\bar{q}, \bar{q}') = \int \mathcal{D}[\bar{q}] e^{-S_E[\bar{q}]/\hbar - \phi_E[\bar{q}]/\hbar}$$

(8)

with the bare Euclidian action $S_E[\bar{q}] = \int_0^{h \beta} d\tau [M \dot{\bar{q}}^2 / 2 + V(\bar{q})]$ of a particle of mass $M$ in the potential $V(\bar{q})$ and the Euclidian influence functional $\phi_E[\bar{q}] = \int_0^{h \beta} d\tau \int_0^{h \beta} d\sigma K(\imath \tau - i \sigma) \bar{q}(\tau) \bar{q}(\sigma)$. To solve the path integral we put $\bar{q} = \bar{q}_{ma} + \delta \bar{q}$, where $\bar{q}_{ma}(\sigma)$ is the minimal action path to (8) and obeys the boundary conditions $\bar{q}(0) = \bar{q}, \bar{q}(h \beta) = \bar{q}'$, while $\delta \bar{q}(\sigma)$ denote quantum fluctuations with $\delta \bar{q}(0) = \delta \bar{q}(h \beta) = 0$. The most convenient way to calculate $\bar{q}_{ma}$ is to switch to
Fig. 1 – $\Lambda$ as a measure for position fluctuations and $\Omega = \langle p^2 \rangle$ vs. inverse temperature for different $\gamma$ and a Drude model with $\omega_c/\omega_0 = 50$. $\Lambda$ is scaled by $\bar{h}/M\omega_0$, $\Omega$ by $\hbar M\omega_0$.

Fourier space with respect to the Matsubara frequencies $\nu_n = n2\pi/\hbar\beta$. In the corresponding equation of motion to leading order the friction term prevails and one finds $\bar{q}_{\text{ma}} = \bar{r}$ (recall that $\bar{x}$ is assumed to be small, namely, of order $1/\sqrt{\gamma}$). Dynamical contributions, however, must be taken into account if we are interested in next-order corrections. The final result for $\bar{q}_{\text{ma}}$ including corrections of order $1/\gamma$ is lengthy and thus omitted here; the minimal action takes the form

$$S_E(\bar{r}, \bar{x})/\hbar = \beta V(\bar{r}) - \Lambda \beta^2 V'(\bar{r})^2 + \frac{\Omega}{2\hbar^2} \bar{x}^2 + O(\Lambda/\gamma) \quad (9)$$

with

$$\Lambda = \frac{2}{M\beta} \sum_{n=1}^{\infty} \frac{1}{\nu_n^2 + \nu_n \gamma(\nu_n)} \quad \text{and} \quad \Omega = \frac{M}{\beta} + \frac{2M}{\beta} \sum_{n=1}^{\infty} \frac{\hat{\gamma}(\nu_n)}{\nu_n + \hat{\gamma}(\nu_n)} \quad (10)$$

Apparently, $\Lambda$ measures the typical strength of quantum fluctuations in position space, while $\Omega$ is via the identification $\bar{x}/\hbar \rightarrow p$ associated with the variance in momentum $\Omega = \langle p^2 \rangle$. In case of Drude damping with a high-frequency cut-off $\omega_c$ both $\Lambda$ and $\Omega$ can be expressed in terms of $\Psi$ functions. Then, for high temperatures $\gamma\hbar\beta \ll 1$ we find $\Lambda \approx \hbar^2 \beta/12M$ and $\Omega \approx M/\beta$. The friction dependence appears as a genuine quantum effect for lower temperatures and for $\gamma\hbar\beta \gg 1$ one has $\Lambda \approx (\hbar/M\gamma\pi) \log(\gamma \hbar \beta/2\pi)$ and $\Omega \approx (M\hbar\gamma/\pi) \log(\omega_c/\gamma)$. With increasing $\gamma$ the strong squeezing of quantum fluctuations in position induces enhanced fluctuations in the momentum (see fig. 1), thus suppressing non-diagonal elements in the density matrix.

By expanding the full action in (8) up to second order in $\delta \bar{q}$, the contribution of quantum fluctuations to (8) is obtained. Due to the strong friction higher-order terms are negligible. The corresponding Gaussian integral is again calculated by utilizing a Fourier expansion in the Matsubara frequencies. Eventually, the equilibrium density matrix in the strong-friction limit is found as

$$\rho_\beta(\bar{x}, \bar{r}) = \frac{1}{Z} e^{-\beta V(\bar{r}) - \Omega \bar{x}^2/2\hbar^2} e^{\Lambda \beta [\beta V'(\bar{r})^2/2 - 3V''(\bar{r})/2]} \quad (11)$$

where $Z$ denotes a proper normalization factor, e.g. $Z = \int dq \rho_\beta(0, q)$. Interestingly, the probability distribution is Gaussian in $\bar{x}$, i.e. its Wigner transform ($\bar{x}/\hbar \rightarrow p$) Gaussian in momentum, even at low temperatures. Anharmonic corrections in $\bar{x}$ to the exponent are at most of order $1/\gamma^2$. The expression (11) is an important result since numerically exact calculations of the path integral (8), e.g. via Monte Carlo techniques, become more expensive at
lower temperatures. It further reveals that for strong friction the equilibrium density consists of a part which in phase space takes the form of a classical distribution, however, with an $h$-dependent $\langle p^2 \rangle$ and a part with $\Lambda$-dependent quantum corrections (see also fig. 1).

Quantum Fokker-Planck equation. – For the time evolution of the density we basically apply the same kind of semiclassical analysis as briefly outlined above. Since on the coarse-grained time scale the dynamics is effectively Markovian, it suffices to calculate the propagating function $J(\cdot)$ for a time step from $t$ to $t + \delta t$, where $\delta t$ obeys $\gamma/\omega_0 \gg \delta t \gg h\beta, 1/\gamma, 1/\omega_c$. Our goal is to derive from this result a time evolution equation for the density which after a Wigner transform gives rise to a quantum Fokker-Planck equation.

We start by expanding the effective action up to second order in $\bar{x}$ and $x(s)$. Specifically, for the potential terms in the real time actions one writes $V(r + x/2) - V(r - x/2) = V'(r)x + O(x^3)$. The complication that arises in evaluating the minimal action paths is that due to the correlations between system and bath the corresponding equations of motions are coupled. For anharmonic potential fields this necessitates in general a numerical evaluation. However, here it turns out that one may write $\varphi_{ma} = r_i + \delta \bar{q}$ and $\varphi_{ma} = r_i + \delta r$, where $\delta \bar{q}$ and $\delta r$ are of order $\Lambda$ or smaller. This way, $\delta \bar{q}$ is determined similarly as for the static case. By approximating $V'(r) = V'(r_1) + V''(r_1) \delta r + O(\delta r^2)$, the real-time paths $\delta r$ and $x$ run effectively in a harmonic oscillator potential with frequency $\sqrt{V''(r_1)/M}$ subject to an external force $V'(r_1)$. The corresponding part of the propagating function has been derived in [11] to which we refer for further details. Hence, to gain a time evolution equation for $\rho(r_1, x_i, t)$ in the form $\dot{\rho}(t) = \mathcal{L} \rho(t)$ we look for an operator $\mathcal{L} = \mathcal{L}(x_i, r_1, \partial/\partial x_i, \partial/\partial r_1, t)$ being at most second order in the coordinates and derivatives. Strong friction forces forward and backward real-time paths to run very close to each other so that quantum fluctuations — responsible for diffusion— reduce to Gaussian noise. Below we will briefly discuss higher-order corrections. To proceed, we make a general ansatz for $\mathcal{L}$ with coefficients specified by comparing $\dot{\rho}$ with $\mathcal{L} \rho$. Eventually, we switch to classical phase space $\{x_i, r_1\} \rightarrow \{p, q\}$, i.e. $\rho(x_i, r_1, t) \rightarrow W(p, q, t)$, by the replacement $x_i \rightarrow i\hbar \partial/\partial p$, $r_1 \rightarrow q$ and $\partial/\partial x_i \rightarrow ip/\hbar$, $\partial/\partial r_1 \rightarrow \partial/\partial q$. This leads to the main result of this article, namely, the time evolution equation for the phase space distribution $W(p, q, t)$ of a dissipative quantum system in the strong-friction limit (7):

$$
\frac{\partial}{\partial t} W(p, q, t) = \left\{ \frac{\partial}{\partial p} [V_{\text{eff}}(q) + \gamma p] - \frac{p}{M} \frac{\partial}{\partial q} + \gamma \langle p^2 \rangle \frac{\partial^2}{\partial p^2} + \frac{\partial^2}{\partial q \partial p} \left[ \frac{1}{\beta} + \Lambda V''(q) - \langle p^2 \rangle /M \right] \right\} W(p, q, t). \tag{12}
$$

Here, we have introduced an effective potential $V_{\text{eff}} = V + (\Lambda/2)V''$, and $\Lambda$ and also $\langle p^2 \rangle = \Omega$ are specified in (10). The first line on the r.h.s. coincides with a classical Fokker-Planck operator in an effective force field [5], the second line describes quantum-mechanical coupled $p$-$q$ diffusion. In the high-temperature limit $\gamma h \beta \rightarrow 0$ the quantum Fokker-Planck equation (QFP) tends to the classical Kramers equation [5]. For small but finite $\gamma h \beta$ and in case of a harmonic potential, the QFP coincides with the master equation gained by Haake and Reibold [12], but differs from the Caldeira-Leggett master equation [13] by the $p$-$q$ diffusion term [14]. However, while these known master equations are restricted (for $\gamma/\omega_0 > 1$) to the range of small $\gamma h \beta$, the new QFP is valid for all $\gamma h \beta$. We only mention here that the above QFP is not of Lindblad form due to the coarse-graining procedure on which its derivation is based. Of course, the equilibrium solution to (12) is given by the Wigner transform of (11). Let us briefly touch the question about higher-order diffusion terms to (12). For harmonic systems they do not occur, so that the QFP is in this sense exact [15]. In case of anharmonic potentials...
Fig. 2 – Relaxation of position expectation values for a damped harmonic oscillator within a Drude model $\omega_c/\omega_0 = 50$, $\gamma/\omega_0 = 5$, $\omega_0 \hbar \beta = 1$. Initial values are for left panel: $\langle p(0) \rangle / M \omega_0 \langle q(0) \rangle = 1$; for right panel: $\langle q^2(0) \rangle = \langle p(0)^2 \rangle / M^2 \omega_0^2 = \langle q^2 \rangle / 4$ (other moments are zero), where $\langle q^2 \rangle$ is the quantum equilibrium variance.

they result from non-Gaussian quantum fluctuations attributed to higher-than-second–order derivatives in $V(q)$. A rough estimate shows that anharmonic terms in $x_f$ (leading to higher-than-second–order derivatives in $p$) in the crucial low-temperature range $\gamma \hbar \beta \gg 1$ are of order $1/[(\hbar \gamma^3/2 \log(\omega_c/\gamma))]$ compared to the leading terms.

Applications. – With the QFP at hand, we are now able to study phase space properties of quantum systems in strongly condensed phase. In particular, classically, due to the time scale separation the strong-friction limit allows for a reduction of the phase space Kramers equation to the position space Smoluchowski equation [5,16] which has been of great importance in a variety of systems in physics and chemistry (cf. the Introduction). Its generalization to the low-temperature quantum domain has been found recently in [8] by focusing on the path integral expression for the diagonal part $\rho(q,q,t)$. Now, from the quantum phase space dynamics this latter result can be rederived and especially the influence of inertia effects can be explored. For this purpose we employ the projection operator techniques invoked in [16] to systematically reduce the QFP to position space. Along these lines we introduce the operators $\mathcal{P} = f_\beta(p) \int dp$ and $\mathcal{Q} = 1 - \mathcal{P}$, where $f_\beta(p)$ is the normalized momentum distribution in equilibrium according to (11). The next steps are straightforward and not presented here in detail. After some algebra one arrives to order $\Lambda/\gamma^3$ at an equation for the position distribution $n(q,t) = f_\beta^{-1} \mathcal{P} W$ of the form

$$\frac{\partial}{\partial t} n(q,t) = \frac{1}{\gamma M} \frac{\partial}{\partial q} \left\{ 1 + \frac{1}{M \gamma^2} [V''(q) + \Lambda V'''(q)] \right\} \mathcal{L}_{QSE} n(q,t). \quad (13)$$

$\mathcal{L}_{QSE} = V'(q) + \partial/\partial q[1/\beta + \Lambda V''(q)]$ is the quantum Smoluchowski operator already derived in [8] (slightly generalized to all values of $\gamma \hbar \beta$). A classical ($\Lambda = 0$) inertia correction $\propto V''(q)$ appears, while quantum fluctuations enter through third- and fourth-order derivatives of the potential. Overdamped quantum Brownian motion in position space thus becomes much more sensitive to the details of the potential profile at lower temperatures.

There is one non-trivial case where analytical results are available and can be compared with exact ones, namely, the damped harmonic oscillator. As already mentioned above, in the quantum case initial correlations between system and reservoir are of crucial importance and
render the calculation even for the harmonic case quite cumbersome [11]. This is particularly true for the strong-friction range where factorizing initial states cannot be used. We illustrate this in detail by considering the relaxation of expectation values in position \( \langle q(t) \rangle \) and \( \langle q(t)^2 \rangle \) (see fig. 2). Already for a moderate damping is the QFP solution for the mean position in very good agreement with the exact dynamics, while that based on a factorizing initial state is completely off. Upon closer inspection one finds that in the latter case the \( q(0) \)-dependence is suppressed by a factor \( 1/\gamma^2 \) compared to the former ones due to the lack of initial correlations. For the mean square in position deviations between the QFP result and the exact one are almost invisible. Remarkably, the impact of quantum fluctuations due to \( \Lambda \) and \( \langle p^2 \rangle \), in the QFP encoded in the \( q-p \) diffusion term, is quite substantial.

**Conclusion.** – We have analyzed the time evolution of strongly damped quantum systems in phase space starting from the exact path integral expression in position space. A suppression of quantum fluctuations in position is associated with enhanced fluctuations in momentum. Our central result is a quantum Fokker-Planck equation applicable from the classical high-temperature (\( \gamma \hbar \beta \ll 1 \)) to the low-temperature quantum (\( \gamma \hbar \beta \gg 1 \)) range. This way, while in the weak-friction limit master equations have been known for years, here, we have given the missing complement for strong damping. The QFP now opens the door to explore phase space features of many systems well studied in classical physics also at lower temperatures, e.g. Kramers rate theory, driven transport, or soft-matter problems.

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