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Microscopic theory of energy dissipation and decoherence in open systems: a quantum Fermi’s golden rule

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Abstract. We shall revisit the conventional adiabatic or Markov approximation, which — contrary to the semiclassical case — does not preserve the positive-definite character of the corresponding density matrix, thus leading to highly non-physical results. To overcome this serious limitation, originally addressed by Davies and co-workers almost three decades ago, we shall propose an alternative more general adiabatic procedure, able to provide a reliable/robust treatment of energy-dissipation and dephasing processes in electronic quantum devices. Unlike standard master-equation formulations, our procedure guarantees a positive evolution for a variety of physical subsystem (including the common partial trace), and quantum scattering rates are well defined even for subsystems with internal structure/ continuous energy spectrum. We shall compare the proposed Markov dissipation model with the conventional one also through basic simulations of energy-relaxation versus decoherence channels in prototypical semiconductor nanodevices.

Present-day technology pushes device dimensions toward limits where the traditional semiclassical or Boltzmann theory \cite{1} can no longer be applied, and more rigorous quantum-kinetic approaches are imperative \cite{2,3}. However, in spite of the quantum-mechanical nature of electron and photon dynamics in the core region of typical solid-state nanodevices \cite{4,5}, the overall behavior of such quantum systems is often governed by a complex interplay between phase coherence and energy relaxation/dephasing \cite{6}, the latter being also due to the presence of spatial boundaries \cite{7,8}. Therefore, a proper treatment of such novel nanoscale devices requires a theoretical modeling able to properly account for both coherent and incoherent — i.e., phase-breaking — processes on the same footing.

To this aim, a quantum generalization of the standard Boltzmann collision operator \cite{9} has been proposed \cite{6}, which however does not preserve the positive-definite character of the density-matrix \cite{9}. Actually, the physical extent to which positiveness is lost was not at all clear in \cite{9}, and thus the standard Boltzmann operator has survived in the semiconductor community, up to now: we shall clearly show, through some basic simulation for prototypical semiconductor nanodevices, that the problem is indeed far more serious than one may suspect.

The only available alternative that guarantees positivity was first formalized by Davies \cite{10,11}, but its range of application is far narrower than the conventional markovian procedure cited above, as it only applies to quantum devices without internal structure (with discrete
spectrum, that is, quantum dots), while it fails to describe relaxation/decoherence in infinitely extended/continuous devices (as it is the case of quantum transport [8]).

Aim of the present Letter is to propose an alternative and more general adiabatic procedure which (i) in the discrete-spectrum case reduces to Davies’ model [10], (ii) for diagonal states gives the well known Fermi’s golden rule [12, 13], and (iii) always describes a Lindblad (and thus positive) evolution, even for continuous energy spectra, thus providing a reliable/robust treatment of energy-dissipation and dephasing processes in semiconductor quantum devices.

Our approximation scheme holds true under the same validity regime of the conventional Markov approach: the so called weak-coupling limit [14], where the subsystem density matrix in the interaction frame moves slowly with respect of perturbative effects.

In order to recall the main features of the problem, let us consider its general formulation based on the fully operatorial approach proposed in [15]. Given a generic physical observable $A$ —described by the operator $\hat{A}$— its quantum plus statistical average value is given by $A = \text{tr} \{ \hat{A} \hat{\rho} \}$, where $\hat{\rho}$ is the so-called density-matrix operator. Its time evolution is dictated by the total (system plus environment) Hamiltonian, that can be regarded as the sum of a noninteracting (system plus environment) contribution plus a system-environment coupling term: $\hat{H} = \hat{H}_0 + \hat{H}^\prime$; the corresponding equation of motion for the density-matrix operator —also known as Liouville-von Neumann equation— in the interaction picture is given by:

$$\frac{d\hat{\rho}^i}{dt}(t) = -i \left[ \hat{\mathcal{H}}^i(t), \hat{\rho}^i \right],$$

(1)

where $\hat{\mathcal{H}}^i$ denotes the interaction Hamiltonian $\hat{H}^\prime$ written in units of $\hbar$.

The key idea beyond any perturbation approach is that the effect of the interaction Hamiltonian $\hat{H}^\prime$ is “small” compared to the free evolution dictated by the noninteracting Hamiltonian $\hat{H}_0$. Following this spirit, by formally integrating Eq. (1) from $t_0$ to the current time $t$, and inserting such formal solution for $\hat{\rho}^i(t)$ on the right-hand side of Eq. (1), we obtain an integro-differential equation of the form:

$$\frac{d\hat{\rho}^i}{dt}(t) = -i \left[ \hat{\mathcal{H}}^i(t), \hat{\rho}^i(t_0) \right] - \int_{t_0}^{t} dt' \left[ \hat{\mathcal{H}}^i(t), \left[ \hat{\mathcal{H}}^i(t'), \hat{\rho}^i(t') \right] \right].$$

(2)

We stress that so far no approximation has been introduced: Eqs (1) and (2) are fully equivalent, we have just isolated the first-order contribution from the full time evolution in Eq. (1).

Let us now focus on the time integral in Eq. (2). Here, the two quantities to be integrated over $t'$ are the interaction Hamiltonian $\hat{\mathcal{H}}^i$ and the density-matrix operator $\hat{\rho}^i$. In the spirit of the perturbation approach previously recalled, the time variation of $\hat{\rho}^i$ can be considered adiabatically slow compared to that of the Hamiltonian $\hat{\mathcal{H}}$ written in the interaction picture, i.e., $\hat{\mathcal{H}}^i(t') = \hat{U}^i(t') \hat{H} \hat{U}^\dagger(t')$; indeed, the latter exhibits rapid oscillations due to the noninteracting evolution operator $\hat{U}_0(t) = e^{-\frac{i\hat{H}_0 t}{\hbar}}$. Therefore, in the standard (and problematic) Markov approximation the density-matrix operator $\hat{\rho}^i$ in interaction frame is simply taken out of the time integral and evaluated at the current time $t$, obtaining an effective equation that (i) has still the double-commutator structure in (2) (and thus conserves the trace, i.e. the total probability can always be normalized to unity); (ii) is local in time (i.e. Markovian); (iii) does not guarantee positivity in general.

Although the last point is of fundamental importance from a physical perspective, it is a common attitude in solid state physics to gauge the problem away, and keep on using the common Markov approximation, by looking at the possible lack of positivity as a transient and negligible effect. As opposed to this, we have performed a simulation of a semiconductor prototypical model, as simple as a two-level quantum dot, interacting with just one resonant
phonon at thermal equilibrium. To be more specific, we chose the quantum dot unperturbed hamiltonian $H_{\text{qd}} = \sum_{\alpha=1}^{2} \omega_{\alpha} c_{\alpha}^{\dagger} c_{\alpha}$, written in units of $\hbar$, where $\omega = \omega_1 - \omega_2 = 7.25$ THz and $c_{\alpha}$ is the annihilation operator for the level $\alpha$. Then the perturbation was taken to be $H' = \sum_{\alpha\beta} a_{\alpha\beta} c_{\alpha}^{\dagger} c_{\beta} b + \text{h.c.}$, where $a_{\alpha\beta}$ are generic coupling parameters, and $b$ is the annihilation operator for the resonant phonon mode with bosonic population $N = (\exp(\hbar \Delta \omega/kT) - 1)^{-1}$, $k$ being the Boltzmann constant and $T$ the absolute temperature. Using the common Markov approximation, we have indeed found the thermal state of the quantum dot as the steady state of the Markov dynamics. But astonishingly enough, we were able to prove that, for quite general choices of the coupling parameters, even a perturbation of the thermal population probabilities as small as $10^{-5}$ could induce exponential divergence of the density matrix eigenvalues: non-negligible lack of positivity is reached within 10 ps (see Fig.1). It is important to stress that these divergences cannot possibly come from numerical rounding errors, as the model has been solved analytically, and only at that point the numerical value of the parameters has been substituted. Moreover, divergences show up only above a critical temperature (computed from the coupling parameters). Both the eigenvalue divergence per se, and its onset at very high temperatures, are clear hallmarks of a non-physical behavior, which in turn forces us to abandon the conventional Markovian approximation for large time modeling of nowadays semiconductor quantum devices.

In order to introduce our alternative formulation of the problem, let us go back to the integro-differential equation (2), and let us consider the following time symmetrization: given the two times $t'$ and $t$, we shall introduce the “average” or “macroscopic” time $T = \frac{t + t'}{2}$ and the “relative” time $\tau = t - t'$. This change of variable has very solid bases, as it is common and well established in a wide variety of contexts, such as Wigner’s phase-space formulation of quantum mechanics [16], standard quantum kinetics Green functions (see e.g. [17]), and even classical radiation theory (e.g., in the treatment of Bremsstrahlung): the basic idea is that the relevant time characterizing/describing our effective system evolution is the macroscopic time $T$. It is now easy to rewrite the second-order contribution in Eq. (2) in terms of these new time variables:

$$\frac{d}{dT} \hat{\rho}^{\dagger}(T) = -\int_{0}^{T} d\tau \left[ \hat{H}^{\dagger} \left( T + \frac{1}{2}\tau \right), \left[ \hat{H}^{\dagger} \left( T - \frac{1}{2}\tau \right), \hat{\rho}^{\dagger} \left( T - \frac{1}{2}\tau \right) \right] \right].$$

Figure 1. Time evolution of the density matrix eigenvalues with initial conditions $\rho_{11}(0) = (1 + 10^{-5}) \frac{N+1}{2N+1}$, $\rho_{22}(0) = 1 - \rho_{11}(0)$ and $\rho_{12}(0) = \rho_{21}(0)^{\dagger} = 0$ (a very small perturbation of the thermal state). Parameters are $\Delta \omega = 7.25$ THz, $a_{11} = 1/20$, $a_{22} = -1/20$, $a_{21} = 1/20\sqrt{2}$, $a_{12} = \sqrt{1/2 - 100a_{21}^{2}/10}$, $N = 100$. Horizontal gridlines define the physical region.

In the spirit of the adiabatic approximation previously recalled, the density-matrix operator $\hat{\rho}^{\dagger}$ can be taken out of the time integral and evaluated at the current time $T$, so that $\hat{\rho}^{\dagger} \left( T - \frac{1}{2}\tau \right) \rightarrow \hat{\rho}^{\dagger}(T)$. As already stressed, this important approximation is valid when the density matrix varies slowly in interaction picture, that is, in the limit of weak-coupling. It is now convenient to replace the finite-domain time integration over $\tau$ by introducing a corresponding Gaussian correlation function $\exp\left\{-\frac{\tau^{2}}{2\tau_{0}^{2}}\right\}$ whose width $\tau_{0}$ may be regarded as a safe overestimation
of the so-called “correlation time”, which is shorter than the period of the system evolution; indeed, for $t - t_0$ greater than the correlation time, the time integration may be safely extended up to infinity. To compare with the exact dynamics in the weak-coupling limit, the correlation time has to finally be brought back to infinity: this in turn may be accomplished by a scaling property of the form $\bar{T}(g) \sim g^{-\xi} T$, for $g \sim 0$, where $g$ is the coupling constant, $\bar{T}$ is a fixed reference time, and $\xi > 0$. Thus in brief, (i) we assume that the density matrix moves slowly with respect to the correlation time $\bar{T}$; (ii) we insert the gaussian weight $\exp\{-\frac{\tau^2}{2\bar{T}^2}\}$ inside the time integral in Eq.(3); (iii) we put $t - t_0 \to +\infty$ for the upper boundary of the latter time integral.

The second crucial step in order to get a genuine Lindblad superoperator for the global dynamics is to exploit once again the slowly-varying character of the density-matrix operator $\hat{\rho}$. The key idea is to perform on both sides of Eq.(3) a set of steps introduced in [b] by the possibly generalized $Q$ eigenvectors of $\hat{H}$.

The above equation can be rewritten in the following compact form:

$$\frac{d\hat{\rho}}{dT} = -\frac{1}{\sqrt{2\pi T}} \int dT' e^{-\frac{T'^2}{2T}} \int d\tau e^{-\frac{\tau^2}{2\bar{T}^2}} \left[ \hat{H}^\dagger \left(T + T' + \frac{\tau}{2}\right), \hat{H}^\dagger \left(T + T' - \frac{\tau}{2}\right), \hat{\rho}(T) \right].$$

Moving back to the original Schrödinger picture and combining the two gaussian distributions, the above equation can be rewritten in the following compact form:

$$\frac{d\hat{\rho}}{dT} = -\frac{1}{2} \left[ \hat{\mathcal{L}}, [\hat{\mathcal{L}}, \hat{\rho}] \right] \quad \text{with} \quad \hat{\mathcal{L}} = \left(\frac{2}{\pi T^2}\right)^{\frac{1}{2}} \int_{-\infty}^{\infty} dt' \hat{H}^\dagger(t') e^{-\frac{t'^2}{2T^2}}.$$  

This is the genuine Lindblad-like superoperator we were looking for; indeed, the operators $\mathcal{L}$ are always Hermitian, and such effective dynamics is positive-definite. We stress how the proposed time symmetrization gives rise to a fully symmetric superoperator, compared to the strongly asymmetric Markov superoperator in [14].

Let us finally rewrite the new Markov superoperator (5) in our noninteracting basis $\lambda$, defined by the (possibly generalized) eigenvectors of $H$: we obtain an effective equation of motion of the form

$$\frac{d\rho_{\lambda_1 \lambda_2}}{dt} = \frac{1}{2} \sum_{\lambda_1' \lambda_2'} \left[ \mathcal{P}_{\lambda_1 \lambda_2, \lambda_1' \lambda_2'} \rho_{\lambda_1' \lambda_2'} - \mathcal{P}_{\lambda_1 \lambda_2, \lambda_1 \lambda_1'} \rho_{\lambda_1 \lambda_1'} \right] + \text{H.c.}$$

with symmetrized quantum scattering rates

$$\mathcal{P}_{\lambda_1 \lambda_2, \lambda_1' \lambda_2'} = \frac{2\pi}{\hbar} H^\lambda_{1 \lambda_1} H^\lambda_{2 \lambda_2} \frac{1}{\sqrt{2\pi\epsilon}} \exp \left\{ -\frac{(\epsilon_{\lambda_1} - \epsilon_{\lambda_1'})^2 + (\epsilon_{\lambda_2} - \epsilon_{\lambda_2'})^2}{4\epsilon^2} \right\}$$

substituting the strongly asymmetrical scattering superoperator given by the conventional Markov approximation [18]. Here, $\hbar$ has been shown explicitly, and $\bar{T} = \frac{\hbar}{T}$ is a measure of the energy uncertainty in the interaction process induced by our temporal coarse graining.

The above scattering superoperator can be regarded as a generalization of the conventional Fermi’s golden rule to the density matrix formalism; indeed, in the semiclassical diagonal case ($\lambda_1 = \lambda_2, \lambda_1' = \lambda_2'$) the above scattering superoperator boils down to what could be considered a dressed vertex-smoothed version of the Fermi’s Golden Rule

$$P_{\lambda \lambda'} = \mathcal{P}_{\lambda \lambda, \lambda \lambda'} = \frac{2\pi}{\hbar} |H_{\lambda \lambda'}^\prime|^2 \frac{1}{\sqrt{2\pi\epsilon}} \exp \left\{ -\frac{(\epsilon_{\lambda} - \epsilon_{\lambda'})^2}{2\epsilon^2} \right\}.$$
and in the limit of infinite correlation-time ($\tau \to 0$) the standard Fermi's Golden Rule [19] is readily recovered.

In passing, we note that the transition rates (7) could be regarded as a "Quantum" version of the celebrated Fermi's Golden rule. This should not generate confusion: of course the Fermi's Golden Rule transition rates are computed according to quantum mechanical calculations (see [19] among thousands of textbooks), but, once computed, they give rise to the Boltzmann equation, which describes a classical Markov process [20] for classical probabilities. Instead, the transition rates (7) do not describe a classical Markov process, but rather its quantum analog: a so called Quantum Dynamical Semigroup [21] for the full density matrix.

Now, the whole theoretical scheme becomes meaningful and applicable only when a well-defined subsystem of interest is identified (together with a corresponding infinite-dimensional environment), so that its correlation time $\tau$ can be estimated, and our (irreversible) semigroup dynamics can correctly describe the subsystem-projected (but fully reversible [22]) exact Hamiltonian dynamics. As final crucial step, we shall show that our conclusions about positivity remain valid no matter how the subsystem is chosen.

To this end, let us now consider a generic projection $P_0$ subsystem [23] on a subalgebra $\mathcal{X}$ of the observable algebra/global system in Heisenberg picture, which is also a completely positive map [24], and let $\{\hat{V}_\alpha\}$ be its Kraus decomposition [25], so that $P_0A = \sum \hat{V}_\alpha^\dagger \hat{A} \hat{V}_\alpha$. Then one could easily verify that the partial trace over a bath is embodied above as a particular case (see for example the formal approaches in [26]). We now observe that, due to its symmetry, our generator keeps the same form of Eq.(5) also in Heisenberg picture: by projecting the latter with $P_0$, and using the completeness relation $\sum \hat{V}_\alpha^\dagger \hat{V}_\alpha = 1$, one can easily write the form for the subsystem's generator in Schrödinger picture, dual to the projected dynamics on the subalgebra $\mathcal{X}$, obtaining $\frac{d}{dt}\rho = -\frac{i}{2} \sum_{\alpha \beta}[\hat{D}^{\dagger}_{\alpha \beta} \hat{D}_{\alpha \beta}, \rho] + \sum_{\alpha \beta} \tilde{D}_{\alpha \beta} \hat{L} \rho \tilde{D}_{\alpha \beta}^\dagger$. Here the "quantum transition amplitude" operators are given, according to (5), by $\tilde{D}_{\alpha \beta} = \hat{V}_\alpha \hat{L} \hat{V}_\beta$ (note that these operators are indeed $t$-dependent, as can be seen from (5)). This Lindblad form [21] shows indeed that we have obtained the generator of the completely positive Quantum Dynamical Semigroup we were looking for. Moreover, the effort we made to consider this rather abstract class of projection is completely justified, as, for example, it paves the way for a new formalism for Quantum Transport: suppose that $\hat{P}$ is a projection in our Hilbert space that identifies, say, a one-dimensional nano-device, and let $\hat{Q}_l$ and $\hat{Q}_r$ project on the left and right contact respectively (obviously $\hat{P} + \hat{Q}_l + \hat{Q}_r = 1$). Then $P_0 \hat{A} = \hat{P} \hat{A} \hat{P} + \hat{Q}_l \hat{A} \hat{Q}_l + \hat{Q}_r \hat{A} \hat{Q}_r$ does belong to the class of projections we have just studied, but the chosen subsystem does not come from a partial trace, nor does it have finite dimensions or discrete spectral properties: its weak-coupling dynamics needs the full power of our theory, in contrast with the previous ones [10, 14].

At this point a few comments are in order. As discussed extensively in [15], also for the simplest case of a standard two-level system —i.e., a generic quantum bit— the standard Markov superoperator predicts a non-trivial coupling between level population and polarization described by the so-called $T_3$ contributions. In contrast, for a two-level system coupled to its environment, the proposed quantum Fermi's golden rule does not predict any $T_3$ coupling term (they vanish in the infinite correlation time limit $\tau \to 0$), thus providing a rigorous derivation of the well-known and successfully employed $T_1 T_2$ dephasing model [27]. In general fact, one could show that for subsystems with discrete spectra, the limit $\tau \to 0$ reduces to Davies' theory [10]. However, for subsystems with continuous spectra, such limit is not defined, but for all finite collision times $\tilde{\tau} > 0$ the proposed approach gives $\tilde{\tau} \sim T_3$, so $T_3$ contributions are indeed present, but they become less and less important as the collision time $\tilde{\tau}$ is raised, as it must be in the weak-coupling limit, to compare with the exact hamiltonian dynamics (see before).

It is also important to say that the pathological eigenvalue divergences we have outlined in our example, for a two-level quantum dot, are completely cured. This is so because the steady state in our approach can be always be shown to be thermal, in the infinite collision time limit,
for discrete energy spectra and under suitable ergodic coupling hypotheses [10]. Finite collision times would then induce a shift in the thermal steady distribution, which in turn would always remain physically meaningful (due to the Lindblad structure of our scattering operator). A detailed study of the foretold shift goes however beyond the scope of the present paper.

To summarize, we have proposed a new approach to modeling nowadays semiconductor nanodevices, by critically reviewing the standard Markov procedure. Indeed, the latter does not preserve the positive-definite character of the density-matrix operator, thus leading to highly non-physical results. To overcome this serious limitation, we have identified an alternative and more general adiabatic procedure which (i) is physically justified under the same validity restrictions of the conventional Markov approach, (ii) in the semiclassical limit reduces to the standard Fermi’s golden rule, and (iii) describes a genuine Lindblad evolution, thus providing a reliable/robust treatment of energy-dissipation and dephasing in state-of-the-art quantum devices. We stress that our formulation generalizes preexisting theories significantly, as it gives a positive dynamics for a considerably large class of projections, i.e. ways to chose the subsystem, and it is well defined even for infinitely extended subsystems (i.e. with continuous spectrum). In turn, this allows to investigate subsystems with both discrete and continuous spectra, a feature largely shared by mesoscale electronic and opto-electronic quantum devices; on the other side, it suggests a new way to treat electrical contacts for infinitely extended quantum devices, thus opening up the exciting possibility of a new formalism for Quantum Transport.

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