A "Square-root" Method for the Density Matrix and its Applications to Lindblad Operators

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June 26, 2021

Abstract

The evolution of open systems, subject to both Hamiltonian and dissipative forces, is studied by writing the \( nm \) element of the time \((t)\) dependent density matrix in the form

\[
\rho_{nm}(t) = \frac{1}{A} \sum_{\alpha=1}^{A} \gamma_n^\alpha(t)\gamma_m^{\alpha*}(t)
\]

The so called "square root factors", the \( \gamma(t) \)'s, are non-square matrices \[ A \] and are averaged over \( A \) systems \((\alpha)\) of the ensemble. This square-root description is exact. Evolution equations are then postulated for the \( \gamma(t) \) factors, such as to reduce to the Lindblad-type evolution equations for the diagonal terms in the density matrix. For the off-diagonal terms they differ from the Lindblad-equations. The "square root factors" \( \gamma(t) \) are not unique and the equations for the \( \gamma(t) \)'s depend on the specific representation chosen. Two criteria can be suggested for fixing the choice of \( \gamma(t) \)'s one is simplicity of the resulting equations and the other has to do with the reduction of the difference between the \( \gamma(t) \) formalism and the Lindblad-equations.

When the method is tested on cases which have been previously treated by other methods, our results agree with them. Examples chosen are (i) molecular systems, such that are either periodically driven
near level degeneracies, for which we calculate the decoherence occurring in multiple Landau-Zener transition, or else when undergoing descent around conical intersections in the potential surfaces, (ii) formal dissipative systems with Lindblad-type operators representing either a non-Markovian process or a two-state system coupled to bosons.

Attractive features of the present factorization method are complete positivity, the no higher than linear increase of the implementation effort with the number of states involved and the introduction of randomness only at the start of the process.

Keywords: Decoherence, Lindblad operators, Landau-Zener crossing, conical intersections, Non-Markovian processes

PACS number(s): 03.65.Bz

1 Introduction

The present authors have proposed a variational formulation to study the time evolution of the density matrix for situations including both Hamiltonian and dissipative processes [1]. This was based on an expression for the time \( t \)-dependent density matrix, originally devised in [2] and developed by [3], which was written formally as

\[
\rho(t) = \gamma(t) \cdot \gamma^+(t)
\]  

In this work we apply this "square root" or "factorization" method to the investigation of quantum trajectories in a decoherent environment.

In the above, the "square-root factors" are the non-square matrix \( \gamma(t) \) whose components are written as

\[
\gamma^\alpha_n(t)
\]  

and its hermitean conjugate. The upper index \( \alpha \) designates the system in the ensemble and the lower index \( n \) the state of the system. In principle, both labels run over infinite values, but for bookkeeping purposes we let \( \alpha \) take \( A \) values and \( n \) take \( N \) values. (Also, in the examples worked out in this paper we have taken for \( A \) and \( N \) finite and small integers.) Thus, \( \gamma \) the primary quantity in the formalism is a rectangular \( N \times A \) matrix. As derived in [4] and in other texts following von Neumann’s method [5], the density matrix is obtained as an ensemble average over all systems. A
The general \( n \) \( m \) component is

\[
\rho_{nm} = \frac{1}{A} \sum_{\alpha=1}^{A} \gamma_{n,\alpha}^{\alpha} \gamma_{m}^{\alpha*}
\]  

(Equation (3) is quoted by numerous quantum mechanics and statistical physics textbooks. The relationship of the \( \gamma \)'s to the wave-functions in an ensemble and other properties of the density matrix are given in the appendix. In equation (1) the dot-symbol is a shorthand for the averaging (a normalized inner product) over \( \alpha \).)

A formally identical form is obtained for \( \rho \) from an alternative definition of the density matrix of an open system, namely by starting with the total density matrix of the system (\( s \)) + its interactive environment (\( e \)) and then taking the trace over the degrees of freedom of the environment [6]. Thus the "factorization" of the density matrix involves no approximation.

To show this, we write a state vector of the combined system as

\[
|\Psi_{s+e}\rangle = \sum_{\alpha} g_{\alpha}^{\alpha} |n_{s}\rangle |\alpha_{e}\rangle
\]

in terms of complete sets of the system and of the environment. From this we form the density matrix operator

\[
\hat{\rho}_{s+e} = |\Psi_{s+e}\rangle \langle \Psi_{s+e}| = \sum_{n\alpha m\beta} g_{\alpha}^{\alpha} g_{\beta}^{\beta*} |n_{s}\rangle \langle \alpha_{e}| m_{s} \langle \beta_{e}|
\]

Tracing over the environment states gives the reduced density matrix for the system alone

\[
\hat{\rho}_{s} = \text{Tr}_{e} \hat{\rho}_{s+e} = \sum_{\lambda} \langle \lambda | \hat{\rho}_{s+e} |\lambda \rangle = \sum_{n\alpha m} g_{\alpha}^{n} g_{\alpha}^{\alpha*} |n_{s}\rangle \langle m_{s}|
\]

The \( n \) \( m \) matrix element of the operator \( \hat{\rho}_{s} \) is

\[
\rho_{nm} = \langle n_{s} | \hat{\rho}_{s} | m_{s} \rangle = \sum_{\alpha} g_{\alpha}^{n} g_{\alpha}^{m*}
\]

which is of the square root form in equation (3) with \( \gamma_{n}^{\alpha} = \sqrt{A} g_{n}^{\alpha} \).

### 1.1 The non uniqueness of square-root factors

The square-root factors are not unique. This can easily be seen from equation (1). Suppose I know a matrix \( \gamma(t) \) and use it to calculate \( \rho(t) \):

\[
\rho(t) = \gamma(t) \cdot \gamma^{\dagger}(t)
\]
Obviously I can take any unitary matrix $U$ in the ensemble space and use it to calculate another matrix $\gamma'(t)$ such that:

$$\gamma'(t) = \gamma(t)U$$

(9)

Or in index notation:

$$\gamma'_{\alpha}^{\beta}(t) = \gamma_{\alpha}^{\beta}(t)U_{\beta}^{\alpha}$$

(10)

Now if $\gamma'(t)$ is used to calculate another density matrix $\rho'(t)$ using equation (1) we obtain:

$$\rho'(t) = \gamma'(t) \cdot \gamma'^{\dagger}(t) = \gamma(t)U \cdot U^{\dagger} \gamma^{\dagger}(t) = \gamma(t)U \cdot U^{\dagger} \gamma^{\dagger}(t) = \rho(t)$$

(11)

Hence $\gamma'(t) = \gamma(t)U$ is just another representation of the density matrix $\rho(t)$, the use of either $\gamma'(t)$ or $\gamma(t)$ can not be distinguished experimentally and has no physical significance. Furthermore we can choose a $U$ matrix evolving in time such that $U = U(t)$. The considerations that dictate which is the best representation to use are discussed in the following sections.

## 2 Evolution in a dissipative environment

### 2.1 A density matrix formulation

At the present the Liouville-von Neumann-Lindblad (LvNL) equations, that are linear in the density matrix and ensure its complete positivity, are frequently employed for the evolution of the density matrix in the presence of dissipative processes. These are written (with the over dot representing time derivation and the time dependence temporarily suppressed) as:

$$\hbar \dot{\rho} = -i[H, \rho] + (2L\rho L^{\dagger} - L^{\dagger}L\rho - \rho L^{\dagger}L)$$

(12)

showing the Hamiltonian ($H$) and dissipative ($L$) processes [7]. (There may be several of the latter, in which case each process is labelled by an index and the rate equation contains a sum over the processes. For notational simplicity we consider a single process and do without an indexed $L$. A theoretical development leading to the above equations is found in [8].) The relation of the Lindblad equation to stochastic (Ito or Stratonovich) formulation of dissipative processes in quantum mechanics was developed in [9], further elucidated in [10] and comparisons between various rate formalism were made in [11]. Several extensions of the Monte-Carlo (MC) or unravelling formalism were made to non-Markovian and other processes (e.g., [12]-[14]). For opposing viewpoints we refer to [15]-[22].
2.2 Time development of the $\gamma$'s

We now postulate time-evolution equations in the "square root" method. The form of the equations is rationalized by the facts that they will correctly give the Liouville-von Neumann equations for Hamiltonian processes and that they have the form of the LvNL equations for the diagonal elements of the density matrix $\rho_{nn}$, when there also dissipative terms. The off-diagonal matrix elements $\rho_{nm}$ ($n \neq m$) will be discussed after introduction of the initial conditions.

\[ h \dot{\gamma}_n^\alpha = -iH_{nr}^\alpha \gamma_r^\alpha + L_{ns}^\alpha \gamma_s^\alpha \gamma_r^\alpha L_{nr}^\alpha (\gamma_s^\alpha)^{-1} - L_{rn}^\alpha L_{rs}^\alpha \gamma_s^\alpha \]  

(13)

The star denotes the complex conjugate. The summation convention for doubly appearing roman indices is used, but $\dot{n}$ in a subscript means no summation over $n$ (and no summation is implied for multiple Greek indices). The inverse $(\gamma_s^\alpha)^{-1}$ is not an element of the inverse matrix of $\gamma$; rather, it is the inverse of an element of $\gamma$. For the conjugate variable one has

\[ h \dot{\gamma}_n^{\alpha*} = i\gamma_r^{\alpha*} H_{rr} + (\gamma_r^{\alpha*})^{-1}L_{rr}^{\alpha*} \gamma_r^{\alpha*} L_{rs}^{\alpha*} \gamma_s^{\alpha*} L_{rn} \]  

(14)

As discussed in previous texts using the square-root method [2, 3], the rate equations for $\rho_{nn}$ follow from combination of equation (13) and equation (14). In fact, using the left hand side of these expressions and carrying out the ensemble averaging, one obtains the time-rate of a diagonal element of the density matrix, as follows

\[ \dot{\rho}_{nn} = \frac{\hbar}{A} \sum_\alpha [\gamma_n^{\alpha*} \dot{\gamma}_n^\alpha + \dot{\gamma}_n^{\alpha*} \gamma_n^\alpha] \]

\[ = \frac{1}{A} \sum_\alpha \{i[\gamma_n^{\alpha*} \gamma_r^{\alpha*} H_{rn} - H_{nr} \gamma_r^{\alpha*} \gamma_n^\alpha] \]

\[ + 2L_{nr}^{\alpha*} \gamma_r^{\alpha*} L_{rs}^{\alpha*} \gamma_s^{\alpha*} L_{rn} - L_{rn}^\alpha L_{rs} \gamma_s^{\alpha*} \gamma_n^\alpha \}

\[ = \{i[\rho, H] + 2L \rho \rho \rho - L \rho L \rho \rho \} \hat{n} \hat{n} \]  

(15)

On the right hand side all products of the $\gamma$'s an be written in terms of density matrix elements and the expression yields precisely the $nn$- component of right hand side of equation (12). Similarly to solutions of the LvNL equations, the trace of $\rho$ is preserved ($\text{Tr} \dot{\rho}(t) = 0$ at all times) and the positivity of any diagonal matrix elements $\rho_{nn}$ is ensured, since $\rho_{nn} = \gamma_n \cdot \gamma_n^* \geq 0$.

On the other hand, the dissipative part of the rate expressions for the non diagonal components of the density matrix has the form

\[ \hbar \dot{\rho}_{nm} = \frac{\hbar}{A} \sum_\alpha [\gamma_n^{\alpha*} \dot{\gamma}_m^\alpha + \dot{\gamma}_n^{\alpha*} \gamma_m^\alpha] \]
\[
\sum_{\alpha} \left\lfloor i \gamma_{\alpha} \gamma_{\alpha}^* H_{\alpha m} - H_{\alpha n} \gamma_{\alpha} \gamma_{\alpha}^* \right\rfloor - \gamma_{\alpha} \gamma_{\alpha}^* L_{\alpha s}^* L_{\alpha r} - L_{\alpha r}^* L_{\alpha s} \gamma_{\alpha} \gamma_{\alpha}^* \\
+ \left. L_{\alpha s} \gamma_{\alpha} \gamma_{\alpha}^* L_{\alpha r}^* \left( \gamma_{\alpha} \gamma_{\alpha}^* \right)^{-1} \gamma_{n} \gamma_{n}^* + L_{\alpha n} \gamma_{\alpha} \gamma_{\alpha}^* L_{\alpha s}^* \left( \gamma_{n} \gamma_{n}^* \right)^{-1} \gamma_{n} \gamma_{n}^* \right\rfloor \\
= \left\lfloor i[\rho, H] - L^1 L \rho - \rho L^1 L \right\rfloor_{nm} + (LB_{\alpha s}^* L^1_{\alpha m})_{\alpha n} + (LB_{\alpha s}^* L^1_{\alpha m})_{\alpha n}
\]

in which the \( B \) tensor is defined as

\[
B_{\alpha s}^{\alpha n} = \frac{1}{A} \sum_{\alpha} \gamma_{\alpha} \gamma_{\alpha}^* (\gamma_{\alpha} \gamma_{\alpha}^*)^{-1} \gamma_{\alpha} \gamma_{\alpha}^*
\]

This contains terms that include the inverse quantities \( (\gamma_{\alpha} \gamma_{\alpha}^*)^{-1} \) and cannot be expressed in terms of the density matrix. This was already noticed in [2]. In the case that \( m = n \) we obtain \( B_{\alpha s}^{\alpha s} = \rho_{rs} \). Thus, while the square root method is self consistent, it is not fully equivalent with Lindblad equations. (A detailed comparison with different methods is given in section 5.) The difference can be clearly formulated by inserting the following expression in the curly brackets in equation (16) and subtracting the same from the last two terms:

\[
L_{\alpha r}^* 2 \frac{\gamma_{\alpha} \gamma_{\alpha}^*}{A} L_{\alpha s} = \left\lfloor 2L^1 \rho L \right\rfloor_{nm}
\]

Then the curly brackets becomes

\[
\left\lfloor i[\rho, H] + 2L^1 \rho L - L^1 L \rho - \rho L^1 L \right\rfloor_{nm}
\]

which is the Lindblad expression, while the difference can be written as

\[
\frac{1}{A} \sum_{\alpha} \gamma_{\alpha}^* (D_{\alpha s}^{\alpha m} + D_{\alpha m}^{\alpha s}) \gamma_{\alpha}^*
\]

with the definition that

\[
D_{\alpha s}^{\alpha m} = (\gamma_{\alpha} \gamma_{\alpha}^*)^{-1} L_{\alpha s} \gamma_{\alpha} L_{\alpha r}^* - L_{\alpha s}^* L_{\alpha r}
\]

The square-root method leads therefore (in general) to different solutions than the Lindblad equation. We repeat that equation (16) is not used to obtain the off-diagonal density matrix terms; rather, these are calculated directly from the square-root factors.
2.2.1 Two components

To complete this subsection we write down explicitly the equations as they should appear for a two component system to be used in the following sections (but suppressing the system index \( \alpha \)):

\[
\begin{align*}
\hbar \gamma_1 & = -i(H_{11}\gamma_1 + H_{12}\gamma_2) \\
& \quad - |L_{21}|^2\gamma_1 - L_{21}^*L_{22}\gamma_2 + L_{12}^*\frac{\gamma_2^*}{\gamma_1}(L_{11}\gamma_1 + L_{12}\gamma_2) \\
\hbar \gamma_2 & = -i(H_{21}\gamma_1 + H_{22}\gamma_2) \\
& \quad - L_{12}^2L_{11}\gamma_1 - |L_{12}|^2\gamma_2 + L_{21}^*\frac{\gamma_1^*}{\gamma_2^*}(L_{21}\gamma_1 + L_{22}\gamma_2)
\end{align*}
\]

\(22\) \(23\)

In the case that the Lindblad operator \( L \) does not have diagonal elements those equations can be further simplified:

\[
\begin{align*}
\hbar \gamma_1 & = -i(H_{11}\gamma_1 + H_{12}\gamma_2) - |L_{21}|^2\gamma_1 + |L_{12}|^2\frac{\gamma_2^*}{\gamma_1} \\
\hbar \gamma_2 & = -i(H_{21}\gamma_1 + H_{22}\gamma_2) - |L_{12}|^2\gamma_2 + |L_{21}|^2\frac{\gamma_1^*}{\gamma_2^*}
\end{align*}
\]

\(24\) \(25\)

2.3 The uniqueness of the \( \gamma \) evolution equation

According to section 1.1 \( \gamma \) is not unique. \( \gamma \) can be replaced by an equally plausible representation \( \gamma' \) such that a unitary matrix \( U \) connects the two:

\[
\gamma_{\alpha}^\alpha(t) = \gamma_{\mu}^{\beta}(t)U_{\beta}^{\alpha}(t)
\]

\(26\)

Taking the derivative of equation (26) we obtain:

\[
\dot{\gamma}_{\alpha}^\alpha(t) = \dot{\gamma}_{\mu}^{\beta}(t)U_{\beta}^{\alpha}(t) + \gamma_{\mu}^{\alpha}(t)\dot{U}_{\mu}^{\beta}(t)
\]

\(27\)

This can also be written as:

\[
\dot{\gamma}_{\alpha}^\alpha(t) = U_{\beta}^{\alpha}(t)[\dot{\gamma}_{\nu}^{\beta}(t) + \gamma_{\mu}^{\beta}(t)\dot{U}_{\nu}^{\beta}(t)(U^{-1}(t))_{\mu}^{\beta}]
\]

\(28\)

Introducing the expression from equation (26) and equation (28) into equation (13) we obtain the result:

\[
\hbar U_{\beta}^{\alpha}(t)[\dot{\gamma}_{\nu}^{\beta}(t) + \gamma_{\mu}^{\beta}(t)\dot{U}_{\nu}^{\beta}(t)(U^{-1}(t))_{\mu}^{\beta}] = U_{\beta}^{\alpha}(t)[-iH_{n\nu}\gamma_{\nu}^{\beta} - L^n_{rs}\gamma_s^{\beta} + L^n_{rs}\gamma_s^{\beta}]
\]

\(29\)
Multiplying by the inverse matrix of $U^{\nu}_{\beta}$ and rearranging terms we obtain:

$$h^{\gamma}_{\gamma} (t) = -iH_{hn} \gamma^t_{\nu} - L_{rn} L_{tn} \gamma^t_{\nu} + L_{n} \gamma^t_{n} L_{hn} U_{\nu} (t) (\gamma^t_{n} U_{\nu} (t))^{-1}$$

$$-h^t_{\nu} (t) U_{\nu} (t) (U^{-1}(t))_{\nu}$$  \hspace{1cm} (30)

We see that the third term in the right hand side of equation (30) (corresponding to the second term in the right hand side of equation (13)) is to some degree arbitrary. Furthermore, if the matrix $U(t)$ is chosen to be time dependent a fourth term can be added without effecting the results. How should one choose a matrix $U(t)$? Some recommendations can be given:

1. Choose a matrix $U(t)$ to simplify equation (30).
2. Choose a matrix $U(t)$ to avoid singularity condition that may occur in the third term in the right hand side of equation (30). More about the problem of singular initial condition in the next subsection.
3. Choose a matrix $U(t)$ in order to reduce the difference between the Lindblad formalism and the factorization matrix formalism. This can be done in terms of the difference tensor $D^{mn\alpha}_{rs}$ defined in equation (21). Inserting the expression from equation (26) into equation (21) we obtain the result:

$$D^{mn\alpha}_{rs}[U_{\beta}] = (\gamma^t_{n})^{-1} L_{n} \gamma^t_{m} L_{n} - L_{sn} L_{rm}$$

$$= (\gamma^t_{n} U_{\nu}(t))^{-1} L_{n} \gamma^t_{m} U_{\nu}(t) L_{n} - L_{sn} L_{rm}$$  \hspace{1cm} (31)

Hence the $U(t)$ can be chosen in order to obtain smaller $D^{mn\alpha}_{rs}$ tensors.

For the examples which were worked out in this paper it was found that choosing the matrix $U(t)$ as the identity matrix produced both simple equations and also good agreement with the Lindblad theory. However, for more involved cases a different choice of the matrix $U(t)$ may be needed.

### 2.4 Initial conditions

The presence of the inverse in the dissipative part of the evolution equations causes the initial conditions (IC) to be of great importance. This is evident, because the rate expressions are singular whenever a component probability is zero. We can simplify the treatment by expressing the density matrix at
$t = 0$ in a diagonal form. This can always be done by a suitable transformation of the basic states. The cases of pure and mixed systems are then easily distinguished. The physical IC are, quite generally, that

$$|\gamma_n^\alpha(0)| = \sqrt{p_n^0} \quad \text{for } n = 1, \ldots, M$$

and zero for the rest of the states. Here the initial probabilities $p_n^0$

$$\sum_{n=1}^{M} p_n^0 = 1$$

by normalization of the density matrix. In a pure state

$$M = 1$$

and in a mixed state

$$M > 1$$

We have already noted that a zero value of a $\gamma$-variable at $t = 0$ (or at any later time), causes a singularity on the right hand side of equation (13) and equation (14). This is overcome by starting the integration at a time arbitrarily close to and slightly above $t = 0$, subject to the IC’s given by

$$\lim_{t \to 0^+} \gamma_n^\alpha(t) = e^{i\phi_n^\alpha} \left( \sqrt{p_n^0} + \sqrt{2|L| p_n^0} e^{i(\phi_n^s - \phi_n^r)} L_n^* t \right)$$

with the phases $\phi_n^\alpha$ taken to be real, but with no other restriction on them. When $p_n^0 \neq 0$, the second term in the above equation can be ignored. But if the first term is zero, the following term is essential. The correction to this term can be shown to be, for small $t$, of the order $|L|^2 t$. It can be checked that the above choice of IC ensures that the singularity on the right hand side of the evolution equation for any component $\gamma$ is exactly cancelled by the singular time derivative on the left hand side. (For a historical remark, singularities of the solutions in the density matrix at $t = 0$ were noted early on, in section 4 of [24]. The fast "slippage" or initially irregular behavior of solutions was recognized in [25].)

We now assume that all solutions of the rate equations for the $\gamma$’s correspond to a physical system in the ensemble. The ensemble averaging has therefore to be carried out for all possible choices of the initial phases.
2.4.1 Short time behavior

Let us now calculate the time development of the density matrix at short times, for an initially pure state, in which

\[ \rho_{11}(t = 0) = 1, \quad \rho_{1n}(0) = 0, \quad \rho_{nm}(0) = 0, \quad \text{for } n \neq 1 \neq m \]  

(37)

In this case

\[ \sqrt{p_{11}^0} = 1 \]  

(38)

and

\[ \sqrt{p_{nn}^0} = 0 \quad \text{for } n \neq 1 \]  

(39)

For the case \( n \neq 1 \) equation (36) takes the form

\[ \lim_{t \to 0^+} \gamma_{n}^\alpha(t) = e^{i\phi_{\alpha,n}} \sqrt{2L_{n1}L_{n1}^*t} = e^{i\phi_{\alpha,n}} |L_{n1}| \sqrt{2t} \]  

(40)

The density matrix has the following short time behavior:

\[ \lim_{t \to 0} \rho_{nm} = \lim_{t \to 0} \frac{1}{A} \sum_\alpha \gamma_{n}^\alpha(t) \gamma_{m}^{\alpha*}(t) \approx g_{nm}(t) \frac{1}{A} \sum_\alpha e^{i(\phi_{\alpha,n} - \phi_{\alpha,m})} \]  

(41)

where

\[ g_{nm}(t) = |L_{m1}| \sqrt{2t} \quad \text{for } n = 1, m > 1 \]

\[ = 2|L_{n1}| |L_{m1}| t \quad \text{for } n \neq 1 \neq m \]  

(42)

The last case includes diagonal matrix elements for the initially absent states, \( n = m > 1 \), but here the phase factor averaging over \( \alpha \) (the system labels) in equation (41) gives unity. These matrix elements are thus seen to give rise to a non-zero value within a time of the order of \( t \approx L^{-2} \).

Due to the dissipative mechanism (represented by \( L \)), the system will therefore become mixed beyond this time, so that \( \text{Tr} \rho^2 < 1 \). This pure-to-mixed transition can also be obtained from calculation of the time derivatives of the square root factors. (With a purely Hamiltonian interaction \( (H) \), an initially pure state for this Hamiltonian will persist to be a pure state at later times.)

3 Decoherence in Molecular Systems

3.1 Driven Level Crossing

The changes in the density matrix involving two states during fast level crossing by a molecular systems were considered in [26] including also dissipative forces. It was later remarked in [27] that the "square root operator
method of [2]”, represents an alternative way of showing how a dissipative
term in the Hamiltonian can cause decoherence. We now formulate the
evolution equations in this square-root scheme and solve the resulting equa-
tions. The solutions for the diagonal term in the density matrix, shown
graphically below in Figure 1 and similar to those shown in [1], have all
the features appearing in the earlier treatments [26, 27] based on entirely
different schemes of solution.

The level crossing (LC) system is characterized by a Hamiltonian (H_{LC})
and a non-Hermitian dissipative (L_{LC}) part written for the two states in
the matrix forms, as follows:

\[
H_{LC} = \left( \begin{array}{cc}
\frac{1}{2}G\cos(\omega t) & J \\
J & -\frac{1}{2}G\cos(\omega t)
\end{array} \right)
\] (43)

\[
L_{LC} = \sqrt{\Gamma} \left( \begin{array}{cc}
0 & 1 \\
1 & 0
\end{array} \right)
\] (44)

G denotes the strength of the periodic driving field; the coefficients J and
\(\Gamma\) of the tunnelling and of the dissipative mechanisms are denoted by same
symbols as in [26] and [27].

For a Markovian process the rate equations are now written for the two
elements (\(\gamma_1^\alpha, \gamma_2^\alpha\)) in the density matrix as

\[
i\hbar \dot{\gamma}_1^\alpha = \frac{1}{2}G\cos(\omega t)\gamma_1^\alpha + J\gamma_2^\alpha - i\Gamma[\gamma_1^\alpha - |\gamma_2^\alpha|^2/\gamma_1^\alpha^*]
\]

\[
i\hbar \dot{\gamma}_2^\alpha = -\frac{1}{2}G\cos(\omega t)\gamma_2^\alpha + J\gamma_1^\alpha - i\Gamma[\gamma_2^\alpha - |\gamma_1^\alpha|^2/\gamma_2^\alpha^*]
\] (45)

We have already called attention to the divisors \(\gamma^*\) on the extreme right,
characteristic of the factorization formalism for dissipative processes, equa-
tion (14). The trace of the density \(\rho(= \gamma \cdot \gamma^+)\) stays constant during the
motion, by construction.

We next solve two equations for the \(\gamma^s\), subject to the pure state initial
conditions \(|\gamma_1^\alpha|^2 = 1, \gamma_2^\alpha \approx 0\) at \(t = 0\). Then from the solutions we form the
diagonal matrix elements of the density matrix and finally show the results
in figure 1. For a beginning, the three lower drawings in the figure arose
from calculations that were carried out for a density matrix referring to an
"ensemble" consisting of one system. This means that one has \(A = 1\) and the
summation over \(\alpha\) in equation (3) is trivial. The quantity changed between
the upper three drawings is the strength \(\Gamma\) of the dissipative term. As this
increases, a transition takes place from the slow to the fast decoherence
regime. We have already noted the remarkable similarity of the results obtained here by the factorization method to those in Figure 1 in [26] and in [27], except that for strong dissipation their drawings show only tiny oscillations, unlike our third drawing from below. In this, drawn for $\frac{\Gamma}{\omega} = 20$, after a very steep initial downward slope (not visible in the figure), both diagonal density matrix elements oscillate about the asymptotic value of $\frac{1}{2}$.

In this trivial "ensemble" of $A = 1$ the off-diagonal matrix element is simply given by $|\rho_{12}| = |\gamma_1 \gamma_2^*| = |\gamma_1||\gamma_2| = \sqrt{\rho_{11}(1 - \rho_{11})}$. This differs from the same quantity calculated and shown in Fig. 2 of [27]. Their curve initially oscillates around 0, and at longer times settles down to this value, whereas our result tends asymptotically to $\frac{1}{2}$. We do not regard this as a serious discrepancy, since in a realistic model, in which $A$ is large, the phase decoherence will make $\rho_{12}$ vanish, as argued in section 2.3.

We have also calculated the density matrices when there is a non-trivial summation, namely when initial conditions are $\gamma_\alpha(0) = e^{i\frac{\pi}{2}}, e^{i\frac{3\pi}{2}}, e^{i\frac{5\pi}{2}}, e^{i\frac{7\pi}{2}}$ for $\alpha = 1,2,3,4$ respectively and $\gamma_2(0) \approx 0$, (so that $A = 4$), instead of having only $\gamma_1(0) = 1$ (and $A = 1$), as before. Physically, this represents a certain type of averaging over the environment. (In more complex systems, one would require an averaging in the density matrix over a much larger number of states, such that $A \to \infty$.) The resultant density tends now to an almost perfectly straight line. This is similar to the graphs shown in both [26] and in [27] for strong dissipation and elucidates the practical consequence of system-averaging in the density matrix. We have also worked out the $A = 4$ case for the lower two graphs in Figure 1. For these graphs, there was hardly any perceptible change from those shown.

For the physical meaning of these results in the context of molecular level crossing, we point to [26] and references therein, while for a more general application to decoherence we refer to [27].

### 3.2 Descent Across a Conical Intersection

Whereas in the previous case the decoherence mechanism was activated by a non-Hermitean dissipative force, whose strength was designated by $\Gamma$, there are situations where dissipation is intrinsic in the dynamics. A situation of this type takes place in the excited state dynamics of a molecular system moving on a potential energy surface as it approaches a conical intersection ($ci$). (This is a frequently encountered natural process, and it has been

\[ \rho_{11} + \rho_{22} = 1 \Rightarrow |\gamma_2| = \sqrt{\rho_{22}} = \sqrt{1 - \rho_{11}} \]
Figure 1: Evolution of a diagonal element of the density matrix $\rho_{11}$. The strength of dissipation increases upwards in the drawings. For all drawings the parameters in equation (45) are chosen as $G = 25, J = 3, \omega = 1$. Then, in the bottom drawing $\Gamma$ (the dissipative parameter) = 0, $A$ (the ensemble size) = 1; in the second drawing (from below): $\Gamma = 0.05, A = 1$; in the third drawing: $\Gamma = 20, A = 1$; in top drawing $\Gamma = 20$ (as in previous, but $A = 4$. The initial downward slope in the top two drawings is too steep to be visible and so are tiny fluctuations in the horizontal part of the top drawing. Note the smoothing effect of the ensemble averaging, evident from a comparison between the top two drawings.
claimed that it is basic to many naturally occurring life processes in which an electronic state is changed, e.g. photosynthesis [28]. ci have been previously studied by numerous researchers, [29] and other papers in that volume [30].) We now briefly give the underlying formal background, with a view of applying to it our formalism. A schematic illustration of a ci is shown in Figure 2.

We first recall the basic result of von Neumann and Wigner [31] that the crossings (points of degeneracy) of potential energy surfaces (=electronic energies as functions of the nuclear coordinates) for a polyatomic molecule can be generally described in terms of just two nuclear displacement parameters. (Following the notation of [32], we shall here denote these by $Q_1$ and $Q_c$, the former being a "tuning" and the latter the "coupling" mode coordinate. They are actually two linearly independent combinations of the nuclear coordinates.) The two surfaces (belonging to the two locally adjacent "adiabatic" electronic states) separate from each other near the intersection point in a manner that is linear in both coordinates: hence the name "conical intersection" (ci). They differ from energy-surface intersections that happen during molecular or atomic collisions, in that these may (approximately) be treated in a single-dimensional coordinate space, usually the separation between the colliding-reacting species. The probability of change of the electronic state in such collisions is given by the Landau-Zener formula [33],[34], which is at the basis of the subject in the previous section. An analogous semi-classical formula for the passage across a ci was obtained by Nikitin [35]. Later developments were summarized in [36]. In a simplified form the expression of [35] for the asymptotic probability of transfer between the two diabatic states in a 2-dimensional space ($x$, $y$) can be written as

$$P_{2 \rightarrow 1}^{\text{diab}} = \exp\left[-\frac{\pi K (v_x^2 + v_y^2) - \frac{1}{2}}{2\hbar l^2}\right]$$

In this formula $v_x$ and $v_y$ are components of the starting velocities on the upper ("2") adiabatic surface, $l$ is the closest distance in the passage to the intersection point and $K$ is the strength of the electron-nucleus dynamic coupling.

The time dependent Schrödinger equation for the dynamics of a ci was subsequently solved numerically in several papers, especially in [37] - [38], which contain references to earlier works. These show that the initial wave packet (which is excited in an upper electronic state) has a definite, non-zero asymptotic probability to end up in the other electronic state as the wave packet traverses the ci. The dynamics bears therefore the irreversibility
Figure 2: Schematic drawings of adiabatic potential surfaces exhibiting conical intersection (ci). The upper and middle drawings show the upper and lower sheets as function of the two displacement coordinates $Q_1$ and $Q_c$ introduced in the text. The lower drawing superimposes these two and shows the locally conical nature of the intersection at $Q_1 = -4$ (expressed in arbitrary units, typically about -0.1 nm) and $Q_c = 0$. The system starts its trajectory on the upper surface as a wave packet centered at (say) $(Q_1 = -10, Q_c = 2)$ (shown by a blob, where it belongs almost entirely to the upper electronic state) and descends towards and beyond the $ci$, oscillating to and forth and (partly) losing its upper electronic state character.
hallmark of a dissipative mechanism, though such mechanism was nowhere introduced in this model (unlike that in the previous section). The irreversibility was explained in [38] as due to the essential anharmonicity of the dynamics in the $Q_1, Q_c$-coordinates. The anharmonicity shows up in the cusps on the energy surfaces at the degeneracy point.

### 3.2.1 A simplified formalism for $ci$

It is customary to represent the two bare or ”diabatic” electronic states (those that are independent of the nuclear coordinates) by the column vectors

$$
\begin{pmatrix}
"1", "2"
\end{pmatrix} = \begin{pmatrix}
1 \\
0
\end{pmatrix}, \begin{pmatrix}
0 \\
1
\end{pmatrix}
$$

(47)

([32]- [39]). Temporarily simplifying the situation of [32] to the case where there is only one tuning mode $Q_1$, as well as the coupling mode $Q_c$, we have for the two coordinates the following harmonic oscillator Hamiltonian:

$$
H_{nuc} = \frac{\hbar \omega_1}{2} (-\frac{\partial^2}{\partial Q_1^2} + Q_1^2) + \frac{\hbar \omega_c}{2} (-\frac{\partial^2}{\partial Q_c^2} + Q_c^2)
$$

(48)

where $\hbar \omega_1$ and $\hbar \omega_c$ are the quanta of vibrational energies. In the doublet representation the nuclear Hamiltonian is written as a scalar or, equivalently, as $H_{nuc}$ times the 2x2 unit matrix $I$.

The nuclear-electronic interaction part can then be written without loss of generality as

$$
H_{el-nuc} = \begin{pmatrix}
\delta E + K_1 Q_1 & K_c Q_c \\
K_c Q_c & -\delta E - K_1 Q_1
\end{pmatrix}
$$

(49)

where $2\delta E$ is the vertical energy offset between the two electronic states and $K_1, K_c$ are the coupling strengths for the two coordinates.

The Schrödinger equation to be solved for the time dependent vector $\Psi(Q_1, Q_c, t)$ is

$$
i\hbar \frac{\partial \Psi(Q_1, Q_c, t)}{\partial t} = (H_{nuc} + H_{el-nuc})\Psi(Q_1, Q_c, t)
$$

(50)

subject to the initial condition at $t = 0$, that $\Psi(Q_1, Q_c, 0)$ is a wave packet (say, of gaussian forms in the two coordinates $Q_1, Q_c$) on the upper potential surface, centered at some position well above the intersection point ($Q_1^i = -\frac{\delta E}{K_1}, Q_c = 0$). This type of wave packet can be conveniently formed.
by short duration (compared to the inverse frequencies $\omega^{-1}_1$ and $\omega^{-1}_c$) optical excitation from some lower lying electronic state. The wave packet will then descend towards the $ci$ point and beyond. Throughout its passage the initially excited (diabatic) electronic state (say, "1") will lose amplitude in favor of the other state "2". This loss will be especially intense near the degeneracy point. Ultimately, for "long" times of the order of picoseconds, the probability will tend to some asymptotic limit (say, $P_{1,\infty}$) between 0 and 1. (Figures 1-4, in [32]. Actually, in any individual run it will slightly oscillate about the asymptotic probability.) This value depends on the parameters of the system and also, presumably to a lesser extent, on the initial conditions.

3.2.2 A Lindblad-type formalism for $ci$

In this section we present a reformulation of the previous two state-two mode system, such that the irreversible nature of the dynamics is built into the model (rather than emerges from the solution). We do this by constructing a phenomenological dissipative term in the square-root formalism, based on the Hermitean formalism of the previous subsection. This procedure will then lead to Lindblad-type terms in the master equations. (Let it be emphasized that we are not deriving the Lindblad term from a microscopic process, but are formulating the microscopic equations in an irreversible setting for the partial, electronic degrees of freedom, having eliminated the nuclear coordinates.)

In the motion of the wave packet on an adiabatic surface the electronic state amplitudes will depend on the position of the nuclear coordinates. In the square root formalism, and relying on Appendix A.1, we write this as

$$\gamma_i^\alpha = \gamma_i^\alpha(Q_1, Q_c) \quad (i = 1, 2)$$  \hspace{1cm} (51)

We invert these relations to make the coordinates some functions of the $\gamma$'s and then linearize the functional relation to be composed of a classical (non-dissipative, real) part and a "dissipative" part which will give rise to a Lindblad type term. Specifically,

$$Q_r(t) = Q_r^{\text{class}}(t) + Q_r^L(t) \quad (r = 1, c)$$  \hspace{1cm} (52)

for either coordinate and then

$$Q_1^{\text{class}}(t) = Q_1^0 \cos(\omega_1 t) + \frac{v_1}{\omega_1} \sin(\omega_1 t)$$  \hspace{1cm} (53)

$$Q_c^{\text{class}}(t) = \frac{v_c}{\omega_c} \sin(\omega_c t)$$  \hspace{1cm} (54)
These relations are appropriate for a classical motion (for which the initial values of the coordinates are \((Q^0, 0)\) and of the velocities are \((v_1, v_c)\); they should provide a fair enough description of the center of a gaussian wave packet. Other choices for the initial values lead to similar results for the asymptotic behavior. (But take note that the level crossing probability is zero when the initial velocities are zero or when the classical pathway goes across the ci. This is evident from Nikitin’s expression for a ci level crossing probability \([35, 36]\), shown above in equation (46).)

The expressions for the Lindblad component of the coordinates are more complicated and their rationale will be apparent only later. They are

\[
Q^L_1(t) = \frac{-i\Gamma}{K_1} \left[ (1 - P_{1,\infty}) |\gamma_1^\alpha|^2 - P_{1,\infty} |\gamma_2^\alpha|^2 \right] \left( \gamma_2^\alpha \gamma_1^{\alpha*} + \gamma_1^\alpha \gamma_2^{\alpha*} \right)
\]

\[
Q^L_c(t) = \frac{i\Gamma}{K_c} \left[ (1 - P_{1,\infty}) |\gamma_1^\alpha|^2 - P_{1,\infty} |\gamma_2^\alpha|^2 \right] \left( \gamma_1^\alpha \gamma_1^{\alpha*} - \gamma_2^\alpha \gamma_2^{\alpha*} \right)
\]

Here \(P_{1,\infty}\) and \(P_{2,\infty}\) are asymptotic weights or probabilities for the two (adiabatic) components. The time dependence of the \(\gamma\)-factors is suppressed in these formula, for brevity. Because of the irreversibility of the process, one can assume the functions \(Q_1, Q_c\) to be complex.

Substitution of the nuclear coordinates, as given in equation (52)–equation (56), into equation (49) and the use of this \(\gamma\)-dependent Hamiltonian in the first (“Hamiltonian”) term in equation (13)–equation (14), yield after some simplification the following Lindblad form of the rate of equations for the \(\gamma\)'s

\[
i\hbar \dot{\gamma}_1^\alpha = \left[ \delta E + K_1(Q_1^0 \cos(\omega_1 t) + \frac{v_1}{\omega_1} \sin(\omega_1 t)) \right] \gamma_1^\alpha + K_c \frac{v_c}{\omega_c} \sin(\omega_c t) \gamma_2^\alpha
\]

\[-i\Gamma \left( 1 - P_{1,\infty} \right) |\gamma_1^\alpha|^2 - P_{1,\infty} |\gamma_2^\alpha|^2 \right] \gamma_1^{\alpha*} \]

\[
i\hbar \dot{\gamma}_2^\alpha = \left[ -\delta E - K_1(Q_1^0 \cos(\omega_1 t) + \frac{v_1}{\omega_1} \sin(\omega_1 t)) \right] \gamma_2^\alpha + K_c \frac{v_c}{\omega_c} \sin(\omega_c t) \gamma_1^\alpha
\]

\[+ i\Gamma \left( 1 - P_{1,\infty} \right) |\gamma_1^\alpha|^2 - P_{1,\infty} |\gamma_2^\alpha|^2 \right] \gamma_2^{\alpha*} \]

The Lindblad operator for this set of equations is:

\[
L_{CI} = \sqrt{\Gamma} \begin{pmatrix} 0 & \sqrt{P_{1,\infty}} \\ \frac{1 - P_{1,\infty}}{\sqrt{1 - P_{1,\infty}}} & 0 \end{pmatrix}
\]
Figure 3: Time development of the weight of the diabatic-state, $|\gamma_1(t)|^2$, following excitation into state "1" and during a passage across a conical intersection with a lower lying state ("2"). Lower curve: a single run. Upper curve (displaced upward by 1): ensemble averaging over four runs ($A = 4$, explained in text). The parameters in equation (57) are (with frequencies and energies /$\hbar$ in inverse femtosecond units, lengths in units of zero point motion amplitudes, typically .05 nanometers): Energy offset, $\delta E = 0.2$, Coupling constants: $K_1 = 0.06$, $K_c = 0.05$. Mode frequencies: $\omega_1 = \omega_c = 0.4$. Initial velocities: $v_1 = 0.24, v_c = 0.32$. Dissipative strength, $\Gamma = -0.1$; Asymptotic weight: $P_{1,\infty} = 0.25$

When at $t = 0$ the "1"-state is excited optically, the initial conditions are $\gamma_1(0) = 1, \gamma_2(0) = 0$ for all $\alpha$, one obtains asymptotically, after some short-time oscillations and rise of the occupation probability of the non-excited "2"-state,

$$\rho_{11}(\infty) = |\gamma_1(\infty)|^2 = P_{1,\infty}, \quad \rho_{22}(\infty) = |\gamma_2(\infty)|^2 = P_{2,\infty} = 1 - P_{1,\infty} \quad (60)$$

The behavior of $\rho_{11}$ as function of time is shown in figure 3. The parameters $K_1, K_c, \Gamma, \omega_1, \omega_c$ are all functions of the parameters in the molecular Hamiltonian (equation (48), equation (49)). $Q_1^0, v_1, v_c$ are defined by the mode of excitation. $P_{1,\infty}$ depends on the molecular Hamiltonian and, as shown in section VI of [38], approximately by the potential surfaces.

It is proper to characterize the present computed case as belonging to the moderately strong dissipative case, since the oscillations about the asymptotic value begin after a time of the order of a vibrational period. A closer look at the curves shows that the computed mean asymptotic line lies above the "input" asymptotic weight $P_{1,\infty} = .25$ by about 0.03. This is an inter-
interesting effect, likely to be due to oscillations in the asymptotic probability which arise from the Hamiltonian part of the rate equation (57). (We call attention to the discussion in section VI of [32], concerning the discrepancy of about the same magnitude and sense between their classical and computed asymptotic probabilities.) Trivially, the other diagonal matrix ($\rho_{22}$) tends to a value that is lower than its asymptotic value, since the two diagonal matrix elements add up to unity. The fluctuations have a roughly uniform period of $2\pi/\omega_1$.

The lower curve is for a single run ($A = 1$), the upper curve (vertically displaced for clarity by unity) is after "ensemble averaging" over four runs ($A = 4$), which differ from each other by having different phases ($\phi_\alpha^0 = 0, \pi/2, \pi, 3\pi/2, \text{ for } \alpha = 1, \ldots, 4$) in the initial $\gamma_\alpha^0(0)$. As seen in Figure 3, no qualitative difference is observed between the curves for a single system and those for the ensemble averaged density matrix, showing that the single system fluctuations due to the periodic Hamiltonian term are not averaged out by the dissipative term.

This last finding changes radically when we apply our formalism to a more complex case, that was proposed in [32] as representative of a $C_2H_4^+$ molecular system. There are now two tuning modes (designated by the subscripts 1 and 2), as well as different frequencies for the three modes. The results for the diagonal density matrix are shown in Figure 4.

The fluctuations are here considerably more congested than in the preceding case, where there was only a single mode-frequency. One sees in Figure 3 that the dissipative mechanism does not completely eliminate the fluctuations. However, additional computations (not shown here) indicate that increasing the dissipation strength $\Gamma$ does further slightly smoothen the intrinsic fluctuations. On the other hand, in the ensemble averaged probabilities (the upper curve in Figure 4) the fluctuations due to the quasi-periodic Hamiltonian are almost completely washed out. We finally note that the computed mean asymptotic value still lies about 0.03 above the nominal asymptotic weight.

4 Some Models of Dissipative Processes

4.1 Non-Markovian processes with memory

Following a work by Diosi et al. [40], we consider the situation in which the Lindblad operator $L$ is dependent on time and hence has the faculty of
Figure 4: Computations in a model for $C_2H_4^+$ (for a single run, the lower curve, and with an "$A = 4$ ensemble averaging" as in the previous figure, the upper curve displaced by unity for clarity). The parameters of Table I (A) and Figure 1 (a) of [32] are used with added values for initial velocities and asymptotic probabilities (chosen somewhat arbitrarily), as follows: $\delta E = 0.95$, Coupling constants: $K_1 = 0.19, K_2 = -0.27, K_c = 0.5$, mode frequencies: $\omega_1 = 0.36, \omega_2 = 0.21, \omega_c = 0.11$, velocities: $v_1 = .6, v_2 = 0, v_c = .8$, $\Gamma = -.1$ (in units as in the previous figure); asymptotic weight: $P_{1,\infty} = 0.25$
Pauli Matrices Expectation Values

Figure 5: Expectation values of Pauli matrices for a two-level system undergoing non-Markovian quantum diffusion (NMQD) for parameters as in Fig. 2(b) of [40] \( w = 1; L = 1; G = 1; W_1 = 1; G_1 = G; a = 0 \). Full line: \( \langle \sigma_z \rangle \); short, broken lines: \( \langle \sigma_x \rangle \); long-broken lines: \( \langle \sigma_y \rangle \).

memory. We consider the simple case of a two-level system, in which:

\[
H = \frac{1}{2} w \sigma_z, \\
L = \sqrt{f(t)} \sigma_- 
\]

The memory function is given by:

\[
f(t) = \frac{G_1}{2} - \frac{\sqrt{G_1^2 - 2G_1l^2}}{2} \tanh \left[ \frac{1}{2} \sqrt{G_1^2 - 2G_1l^2} \right] + \frac{G_1}{\sqrt{G_1^2 - 2G_1l^2}} \tanh \left[ \frac{G_1}{\sqrt{G_1^2 - 2G_1l^2}} \right]
\]

The above Hamiltonian and Lindblad operator lead to the equations:

\[
\dot{\gamma}_1^\alpha = -\frac{1}{2} iw \gamma_1^\alpha - f(t) \gamma_1^\alpha \\
\dot{\gamma}_2^\alpha = \frac{1}{2} iw \gamma_2^\alpha + f(t) \frac{|\gamma_1^\alpha|^2}{\gamma_2^\alpha^*}
\]

The time evolution of the expectation value of the Pauli matrices is plotted in Figure 5 in the case that the above differential equations are solved for the pure state initial conditions \( \gamma_1^\alpha(0) = \frac{3}{\sqrt{10}}, \gamma_2^\alpha(0) = \frac{1}{\sqrt{10}} \) independent of \( \alpha \). The obtained results are very similar to those of [40].
4.2 Probabilities for a two-state system coupled to a bosonic reservoir

This problem was recently studied, alongside with other instances also involving memory, in [41, 42]. The case that we numerically solve using the square-root formalism possesses an exact solution [shown in Eq. 57 of [41]], with which our computed values agree perfectly (Figure 7).

In the model a degenerate two-state system, $|1>$ and $|2>$ (whose Hamiltonian is taken as zero), is coupled to a reservoir of bosons through a time-dependent Hamiltonian interaction term. This is given by

$$H_{int} = B(t)|2><1| + B^*(t)|1><2|$$  \hspace{1cm} (64)

where $B^*(t)$ is a reservoir excitation operator [Eq.(52) in [41]]. The essential quantity in the dynamics is the spectral density given by

$$J(\omega) = \frac{1}{2\pi} \frac{lG_1^2}{(\omega - \omega_0)^2 + G_1^2}$$  \hspace{1cm} (65)

where $l$ is the interaction strength, $G_1$ is the spectral width and $\omega_0$ the characteristic frequency in the reservoir. The analytical solution for the probability of the state occupation $|2>$ that is excited at $t = 0$, is

$$\rho_{22}(t) = e^{-G_1 t}\left[\cosh \frac{\Omega t}{2} + \frac{G_1}{\Omega} \sinh \frac{\Omega t}{2}\right]$$  \hspace{1cm} (66)

where $\Omega = \sqrt{G_1^2 - 2l^2G_1}$.

In our formalism, we solve numerically the master equations for $\gamma_1^\alpha(t)$, $\gamma_2^\alpha(t)$ and their complex conjugates, employing a Lindblad operator $L = f(t)|2><1|$, with the memory function $f(t)$ given in equation (62). Initial conditions are the same for all $\alpha$. The result is shown in Figure 6, which completely overlaps the exact result from equation (66). The Monte Carlo method of [41, 42] also agrees with the analytic results, but uses $10^7$ runs. It is not clear to us what is the minimum runs that is required to obtain agreement, but the expediency of the square root method is evident.

In a further figure we also show the expectation value of the three angular momentum matrices taken with respect to the state amplitudes $(\gamma_1^\alpha(t), \gamma_2^\alpha(t))$. (Figure 7). They are similar to the drawings in Figure 6, but with more persistent oscillations.

\footnote{This means that the ensemble contains only a single system}
Figure 6: Probability of excited state occupancy $\rho_{22}(t) = |\gamma_2^{0}(t)|^2$ against normalized time ($\omega t$). The parameters are $l = 1$, $G_1 = 0.2$. Results are computed by the square root method. They overlap completely the analytic expression.

Figure 7: Averages of Pauli, angular momentum matrices for the solutions against reduced time. $\langle \sigma_z \rangle$: full lines; $\langle \sigma_x \rangle$: long broken lines; $\langle \sigma_y \rangle$: dashed lines. Parameters are as in Fig. 6
5 Comparison with Other Methods.

In this section we briefly consider similarities and differences between the "square-root method" and some other methods, already mentioned in the Introductory section.

(I) Number of independent rate equations.

At any instance of time the density matrix, whether by its definition in equation (3) or by environment tracing, is composed of $N \times A$ independent (complex) numbers ($N$ being the nominal number of states and $A$ either the number of systems in the ensemble or the total number of states in the environment). Thus, in principle, one needs just $N \times A$ rate equations to obtain the density. The square root method postulates $N \times A$ equations. However, the roles of $N$ and $A$ are different and we find that frequently one can get good approximations (to the important diagonal terms) by keeping $A = 1$ or another small number. (e.g., $A = 4$). (Non-diagonal terms may need larger $A$-values)

Rate equation methods based on the density matrix directly work in principle with $\frac{1}{2}N(N + 1)$ equations (cf. [27] and references), which number is for high values of $N$ much larger than that needed in the square root method. The frequently used Monte Carlo or "unravelling" method solves $N \times M$ equations, where $M$ brings in the stochasticity of the environment and can be very large, e.g. 100 in [9], 4096 in [43] and $10^7$ in [41]).

(II) A New Form of the Rate Equations.

The square-root method is formally unlike previously employed methods, such as those surveyed in [11], and it can be reasonably expected not to be reducible to them. To support this claim, we call attention to the inverse "square-root factors" $\gamma^{-1}$ or $(\gamma^+)^{-1}$ appearing in equation (13) and other equations that follow on. (These factors possess an apparent similarity with the "continued fraction method" described in [18] and recently applied in [44], but are essentially different.) The presence of these inverse factors finds expression also in a novel form of the initial conditions, introduced in section 2.1.

(III) Initial time randomization

As has been shown in section 2.1, the environment induced randomization affects the $\gamma$-variables only at the starting time, not during the time evolution. This approach was stressed in two early papers by van Hove [23, 24] in connection with the derivation of the Pauli master equation. In our formulation the initially randomized $\gamma$-s lead, in turn, to the ensemble averaging over various phases in a natural way. Thus the square-root
procedure appears to be self consistent.

In the unravelling method, randomization takes place throughout the time development (namely, by the action of random forces on the system). It seems to us that the ensemble averaging as done in the square root method (namely, by introducing randomness only at the beginning) is closer to the meaning originally ascribed to the density matrix. As asserted at various places in [23], not only is the repeated stochasticity "superfluous", but "it is strictly speaking incompatible with the Schrödinger equation". (We understand that van Hove refers here to the form of the Schrödinger equation as a first order time-dependent differential equation, so that the system’s evolution is fully determined by the initial conditions.)

(IV) Form of the Lindblad operator

As emphasized at several points in this paper, the choice of the Lindblad operator \( L \) in the square root method is determined by the requirement that the evolution equation for the \( \gamma \)'s should lead to rate equations for the diagonal terms in the density matrix, that are formally identical to those in Lindblad’s method [7]. (We assume that this choice is unique.) Thus, we do not derive the \( L \)-operator from an atomic model for the system and its environment. In this sense, the square root method is "phenomenological".

However, most density matrix methods are such, too. As an example, we call attention to the cases studied in the classic paper of [9]. In some works, such as [49] on quantum diffusion, a Lindblad operator was derived from a kinematic model, but only through making some approximations. We have already noted some dissension, [15]-[22], regarding the employment of the Lindblad formalism, especially for initial conditions that are non-separable between the system and its environment [19, 22].

(V) Pure-to- mixed state transition

It has been shown in section 2.1 that in the presence of dissipative processes, a pure state becomes a mixed state in a time of order \( L^{-2} \). This is also the standard result in other formalisms, obtained theoretically, e.g., in [25], and by numerical calculations [27].

(VI) Off-diagonal terms in the density matrix.

For dissipative systems here lies probably the most significant discrepancy between the square-root and density matrix methods. (In a Hamiltonian system, the two formalisms are equivalent, as already noted.) Our defining equations for off-diagonal elements differ from Lindblad’s equations, and therefore so do (in general) the calculated values of the diagonal terms. In a particular case, "level crossing" treated in section 3.1, we have pointed out the discrepancy involved in the square-root procedure. Even in this
case, the calculated behavior of the diagonal element was very similar to that obtained by another method in [27].

It seems that the source of the discrepancy is in the different approximations made to arrive at a dynamical semi-group from a Hamiltonian system, either by truncation of the time domain or by the handling of phase decoherences [7]. So far we have not found any result that would tend to invalidate the present approach, or even to lay bare any shortcomings. In view of the questions surrounding the positivity of linear maps with initially entangled states [21], the circumstance that our rate equations for non-diagonal terms differ from Lindblad’s should not invalidate the square root method.

6 Conclusion

The ”square-root” method (previously used to minimize the action in a general time dependent process [1]) has now been applied to several situations where decoherence is expressible by a Lindblad formalism.

Two irreversible molecular processes (a driven Landau-Zener process and the descent to equilibrium across a conical intersection) were formulated by the square root formalism. Further illustrations of the method were quantum diffusion and memory processes. When compared with results given in the literature and obtained using different procedures, the present method has led in all cases to good agreement and at the cost of very much less effort. The success of the method would seem to justify future uses of the formalism, as for Quantum Brownian Motion, already widely studied in the literature with density matrix methods, [45] -[53] and regarding the question of thermalization [54, 55].

At the same time, since the basic equations in the square root method and other approaches appear to be different in some respects, there remains a theoretical challenge to explain the agreement between the results. Another task is the justification of the basic rate-equation expressions in the formalism by starting from microscopic models.

Appendix
A  The square-root factored density matrix.

A.1 Matrix formulation of square root factors

Though the density matrix in the square root factor form has already appeared before in [2] and [3], we introduce it here following textbook descriptions of von Neumann’s matrix method ([4],[5]). Let $\Psi_\alpha$ be a possible wave function describing the quantum state of the $\alpha$’th system in the ensemble ($\alpha = 1, 2...A$). It can be expanded in terms of an orthonormal set of eigenstates $u_n$ as

$$\Psi_\alpha = \sum_{n=1}^{N} \gamma^\alpha_n u_n \quad (67)$$

Here the size of the orthonormal set (in principle, infinite) is taken as having a finite size $N$. From the normalization of all the wave functions $\Psi_\alpha$ we obtain:

$$1 = \langle \Psi_\alpha | \Psi_\alpha \rangle = \left( \sum_{n=1}^{N} \gamma^\alpha_n u_n \right) \left( \sum_{m=1}^{N} \gamma^\alpha_m u_m \right) = \sum_{n=1}^{N} \sum_{m=1}^{N} \gamma^\alpha_n \gamma^\alpha_m \langle u_n | u_m \rangle = \sum_{n=1}^{N} \sum_{m=1}^{N} \gamma^\alpha_n \gamma^\alpha_m \delta_{nm} = \sum_{n=1}^{N} |\gamma^\alpha_n|^2 \quad (68)$$

As derived in [4] and other texts, the density matrix arises from the ensemble average over all systems in the sense that its $nm$ component is

$$\rho_{nm} = \frac{1}{A} \sum_{\alpha=1}^{A} \gamma^\alpha_n \gamma^\alpha_m \quad (69)$$

The $\gamma^\alpha$’s are rectangular matrices of size $N \times A$, distinct for each $\alpha$ (or system in the ensemble) and the $\gamma^\alpha$’s are conjugate matrices of size $A \times N$. Calculating the trace of the matrix $\rho$ we obtain:

$$\text{Tr} \rho = \sum_{n=1}^{N} \rho_{nn} = \sum_{n=1}^{N} \frac{1}{A} \sum_{\alpha=1}^{A} \gamma^\alpha_n \gamma^\alpha_n = \frac{1}{A} \sum_{\alpha=1}^{A} 1 = 1 \quad (70)$$

for which we have used equation (68).

A.2 Properties of the density matrix

A.2.1 The density matrix eigenvalues

Let us derive some of density matrix well known properties. First $\rho$ is hermitian, since $\rho^\dagger = \rho$, as can be clearly seen from the definition of $\rho$ given
in equation (3). Thus, all eigenvalues $\lambda_i$ of $\rho$ are real which is a property of all hermitian matrices.

Second, the sum of all the eigenvalues is unity. This can easily be shown as follows. There exists a basis of eigenvectors in which $\rho$ is diagonal: in this basis $\rho$ will be denoted by $\rho_D$. The diagonal elements of $\rho_D$ are just the eigenvalues $\lambda_n$. Furthermore, given $\rho$ in an arbitrary basis there exists a unitary matrix $U$ which transforms $\rho$ to $\rho_D$ according to the following equation:

$$\rho_D = U\rho U^\dagger \Rightarrow \rho = U^\dagger \rho_D U$$

(71)

By virtue of equation (70) we obtain:

$$1 = \text{Tr } \rho = \text{Tr } U^\dagger \rho_D U = \text{Tr } \rho_D = \sum_{n=1}^{N} \lambda_n$$

(72)

Third, we will show that all $\lambda_n$ are positive (or zero). To prove this take an arbitrary vector $X$ one can see that:

$$X^\dagger \rho X = \sum_{n=1}^{N} \sum_{m=1}^{N} \frac{1}{A} \sum_{\alpha=1}^{A} X^n_{\alpha} \gamma_{n_{\alpha}} \gamma_{m_{\alpha}}^* X_m$$

(73)

In which the definition of $\rho$ given by equation (3) was utilized. Next define $R^\alpha = \sum_{n=1}^{N} X_{n_{\alpha}}^* \gamma_{n_{\alpha}}$, and obtain the result:

$$X^\dagger \rho X = \frac{1}{A} \sum_{\alpha=1}^{A} |R^\alpha|^2 \geq 0$$

(74)

Further, we write $X$ in the eigenvector basis $V_i$ as $X = \sum_i C_i V_i$ which yields:

$$X^\dagger \rho X = \sum_i C_i^\dagger V_i^\dagger \rho \sum_j C_j V_j = \sum_i C_i^* V_i^\dagger \cdot \sum_j \lambda_j C_j V_j$$

(75)

but since $V_i^\dagger \cdot V_j = \delta_{ij}$ we have:

$$\sum_i \lambda_i |C_i|^2 = X^\dagger \rho X \geq 0$$

(76)

and, since the $|C_i|$'s are arbitrary, we obtain $\lambda_i \geq 0$ for every $i$. From the positivity of $\lambda_i$ and equation (72), we reach the main conclusion of this section, that is:

$$0 \leq \lambda_i \leq 1 \quad \forall i$$

(77)
### A.2.2 Pure and mixed states

From equation (72) and equation (77) one can reach the following classification of density matrices: either there exists a special index $s$ such as $\lambda_s = 1$ while for all $i \neq s$ $\lambda_i = 0$ or that $\lambda_i < 1$ for all indices $i$. The first case is denoted as a "pure" state while the second is denoted as a "mixed" state.

For the pure state we have in the diagonal basis $\rho_D^2 = \rho_D \rho_D = \rho_D$ and also in an arbitrary basis

$$\rho^2 = \rho \rho = U^\dagger \rho_D U U^\dagger \rho_D U = U^\dagger \rho_D U = \rho \quad (78)$$

which is a necessary and sufficient condition for a density matrix to describe a pure state. One obviously obtains also the result

$$\text{Tr} \rho^2 = \text{Tr} \rho = 1 \quad (79)$$

For the mixed state we have

$$\text{Tr} \rho^2 = \text{Tr} \rho_D^2 = \sum \lambda_i^2 < \sum \lambda_i = \text{Tr} \rho = 1 \quad (80)$$

In summary, we conclude that

$$\text{Tr} \rho^2 \leq 1 \quad (81)$$

in which the equality sign is appropriate only in the pure case.

As an example for a pure state take an ensemble for which $A = 1$ and $\rho_{nm} = \gamma_n \gamma_m^*$ in this case:

$$\text{Tr} \rho^2 = \sum_n (\rho \rho)_{nn} = \sum_n \sum_m \rho_{nm} \rho_{mn} = \sum_n \sum_m \gamma_n \gamma_n^* \gamma_m \gamma_m^*$$

$$= \sum_n |\gamma_n|^2 \sum_m |\gamma_m|^2 = 1 \quad (82)$$

Another obvious case of a pure state is an ensemble with an arbitrary number $A$ of wave functions, but in which all the wave functions are equal. Since $\rho_{nm} = \frac{1}{A} \sum_A \gamma_n^* \gamma_m^* = \gamma_n^* \gamma_m^*$ the same argument as above can be applied.

It remains to show that in case that not all the wave functions are equal (in a non-trivial sense) we obtain $\text{Tr} \rho^2 < 1$. 

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A.2.3 Mixed states

Let us calculate the trace of the square density matrix:

$$\text{Tr} \rho^2 = \sum_n \sum_m \rho_{nm} \rho_{mn} = \sum_n \sum_m \frac{1}{A} \sum_{\alpha=1}^{A} \gamma_n^{\alpha} \gamma_m^{\alpha*} \frac{1}{A} \sum_{\beta=1}^{A} \gamma_m^{\beta} \gamma_n^{\beta*}$$  \hspace{1cm} (83)

By defining the "state averaged density function" by:

$$M_{\alpha\beta} = \sum_n \gamma_n^{\alpha} \gamma_n^{\beta*}$$  \hspace{1cm} (84)

we see that:

$$\text{Tr} \rho^2 = \frac{1}{A^2} \sum_{\alpha=1}^{A} \sum_{\beta=1}^{A} |M_{\alpha\beta}|^2$$  \hspace{1cm} (85)

Hence we need to show that for all $\alpha$ and $\beta$, $M_{\alpha\beta} \leq 1$ we shall be particular interested in the case that the inequality is definite that is $M_{\alpha\beta} < 1$.

Let us look at two arbitrary complex vectors: $F, B$ and a complex scalar $\alpha = |\alpha|e^{i\phi}$. Obviously

$$\sum_i |F_i + \alpha B_i|^2 \geq 0$$  \hspace{1cm} (86)

However

$$\sum_i |F_i + \alpha B_i|^2 = \sum_i |F_i|^2 + |\alpha|^2 \sum_i |B_i|^2 + \alpha \sum_i B_i F_i^* + \alpha^* \sum_i B_i^* F_i$$  \hspace{1cm} (87)

Now denote

$$a = \sum_i |B_i|^2, c = \sum_i |F_i|^2$$  \hspace{1cm} (88)

which are both real and positive quantities, and

$$2b = e^{i\phi} \sum_i B_i F_i^* + e^{-i\phi} \sum_i B_i^* F_i$$  \hspace{1cm} (89)

which is a real quantity. And we arrive at the inequality

$$a|\alpha|^2 + 2b|\alpha| + c \geq 0$$  \hspace{1cm} (90)

Since, as function of $|\alpha|$, this is an equation of a parabola which has all its values above the $|\alpha|$ axis (except when the equality holds in this case the parabola touches the axis in a single point), it follows that the discriminant
of this equation is either negative or zero, the latter in the case that
the parabola touches the axis in a single point. Thus we obtain
\[ b^2 \leq ac \]  
(91)

Now assume that the length of the \( F \) and the \( B \) vectors is unity. That is \( a = c = 1 \). Further more denote \( X = \sum_i B_i F_i^* = |X|e^{i\phi} \). Thus \( b \) is equal to
\[ b = |X|\cos(\phi + \varphi) \]  
(92)

The following inequality is obtained
\[ |X|^2 \leq \frac{1}{\cos^2(\phi + \varphi)} \]  
(93)

By choosing the arbitrary phase
\[ \phi = -\varphi \pm n\pi \]  
(94)

we obtain the result
\[ |\sum_i B_i F_i^*|^2 = |X|^2 \leq 1 \]  
(95)

this being a special case of the Cauchy-Schwarz inequality. Next let us
discuss the case in which the equality sign holds in the above equation, that is the case in which \( b^2 = ac \) and the equation \( a|\alpha|^2 + 2b|\alpha| + c = 0 \) is satisfied for a single value of \(|\alpha|\). This can be traced to equation (86) for which we have
\[ \sum_i |F_i + \alpha B_i|^2 = 0 \]  
(96)

but this is only possible for \( F_i = -\alpha B_i \). However, since the length of both
vectors is 1 we arrive at the equation
\[ F_i = -e^{i\varphi} B_i = e^{i(\phi+\pi)} B_i \]  
(97)

Thus in order for the equality to hold the vectors \( F \) and \( B \) must be the same up to a "global" phase. We can now show that \( M_{\alpha\beta} \leq 1 \) and in particular \( M_{\alpha\beta} < 1 \), if \( \gamma^\alpha \) and \( \gamma^\beta \) are different in a "non-trivial" way (a global phase change does not count as a difference). This is done by identifying \( F = \gamma^\alpha, B = \gamma^\beta \). Thus, according to equation (95),
\[ |M_{\alpha\beta}|^2 = |\sum_n \gamma^\alpha_n \gamma^{*\beta}_n| \leq 1 \]  
(98)
and, in particular,
\[ |M_{\alpha\beta}|^2 = |\sum_n \gamma_n^\alpha \gamma_n^\beta| < 1 \] (99)

unless \(\gamma_n^\alpha\) and \(\gamma_n^\beta\) are the same up to a global phase. This yields, barring a unique case,
\[ \text{Tr} \rho^2 = \frac{1}{A^2} \sum_{\alpha=1}^{A} \sum_{\beta=1}^{A} |M_{\alpha\beta}|^2 < 1 \] (100)

Thus a mixed state is really mixed in the sense, that is should contain at least two wave functions which are different in a non trivial way.

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