I. INTRODUCTION

This study aims to illuminate the role of initial conditions in the nonequilibrium dynamics of electrons in small (nanoscopic) quantum systems. This is a continuation of a series of papers devoted to this topic [1–8].

Presently we will discuss several general concepts of quantum transport theory of small systems using a simple non-interacting molecular bridge model as a testing ground. This model covers many aspects of the behavior of many particle systems well, in spite of its simplicity. After inspecting possibilities how to describe a fast initial transient process of the system and to incorporate correlated initial conditions, influencing essentially the systems behavior at the first stage of their evolution, we will concentrate on questions related to decay of the initial correlations and of a simplified description of stages of systems evolution, after initial conditions cease to be important. In particular, we will explore the possibility to describe the nonequilibrium behavior of small systems using asymptotic transport equations, such as General Master Equations (GME) or even Master equations (ME) for single particle distribution functions. These asymptotic equations will be obtained from the equations of motion for Nonequilibrium Green’s functions (NGF) under special assumptions about time evolution of the system.

Double time Nonequilibrium Green Functions (NGF) approach is a natural choice for a proper description of small many body systems out of equilibrium: there are strong interactions in these systems, and so it is easy to bring them far from equilibrium and to induce fast transient processes in them. They are then typically of a quantum nature because of strong coherence phenomena.

Small systems must be open in order to participate in a transport process, which means that the complete system consists of the small system + an extended environment (bath, leads etc.). All degrees of freedom can be divided into two groups: a smaller, often finite group of "relevant" degrees of freedom, and the rest taking care of decoherence, life-time effects, transport phenomena etc.

It might appear as natural to formulate quantum transport theory in terms of reduced density matrix for relevant degrees of freedom [10]. We may briefly summarize the reasons for preferring the use of the complementary approach based on the NGF [1, 5, 11 - 21] as follows:

First, as concerns the one-particle observables, like occupation numbers, currents and current densities, the NGF technique permits to evaluate their non-equilibrium time evolution, just as the density matrix techniques.

Second, due to its double time structure, the NGF approach brings the following advantages: a. it works with renormalized objects in a natural way. This includes the dressed propagators (including quasi-particles and their life-times), on-shell ("E(k)- fixed") as well as off-shell (E and k independent) scattering and renormalized effective interactions,

b. it is possible to formulate its perturbation structure in a self-consistent manner, an important feature for the transport theory,

C. it captures coherences and decoherences in the system in a natural way.

Third, the full double time Green’s function provides a complete description of quantum transport wherever it is necessary to account for the initial correlations or for the quantum coherence, which is a task difficult to achieve in the density matrix framework. At the same time, Green’s functions offer both the criteria and well defined procedures for a controlled transition to the single time quan-
We may now formulate the subject of this paper more precisely: an implementation of the NGF technique in the case of a fast transient process starting at a finite initial time from an initial condition induced by a non-stationary variation of boundary conditions (leaving the less drastic effects of a time-dependent external field aside). We shall first discuss the NGF transport theory with initial correlations on a general level, then go over to its implementation for the molecular bridge model.

Several ways of incorporating complex initial conditions in the NGF formalism have been described and partly used in literature [5, 7, 15, 22–33]. We find it advantageous to attack the initial conditions problem for the real time Green’s functions (Keldysh approach). This permits to employ the so-called time partitioning method introduced in [7] and further developed and used in [5, 6, 8, 9]. The first topic of this contribution will be to show how time partitioning works in the case of the non-interacting molecular bridge model in application to its dynamics under various, both uncorrelated and correlated initial conditions.

The early period of a transient process brought about by an abrupt change of the boundary conditions is characterized by a complex evolution known under the name of decay of initial correlations. Later, as soon as the external conditions change but gradually, the system tends to a kinetic stage as the intermediate and late period of the process. The evolution during the kinetic stage is governed by simplified dynamic equations, quantum transport equations (QTE) for a single-time distribution function of real particles or quasi-particles, generally speaking. This is the contents of the famous Bogolyubov conjecture about the decay of initial correlations [19, 34–36], which has to be verified for each specific system, like, in particular, the molecular bridge.

The second broad topic of the paper is a general scheme of reducing the complete double-time NGF machinery to a QTE. It is based on the so-called Reconstruction theorem [1, 2, 5], according to which the complete particle correlation function and its contraction to the distribution function are constructed in a unified cycle, in which they serve as mutual input in the dynamical equations. A standard tool for shortcutting this cyclic task has been one type of approximate decoupling of the particle correlation function going by name of Kadanoff-Baym Ansatz [1, 11, 18, 19]. This Ansatz has been modified to a class of the so-called causal Ansätze including a particularly successful Generalized Kadanoff-Baym Ansatz [1, 5, 7, 18, 19, 37, 38]. We will use these ansätze to reduce the full NGF description to kinetic equations of the GME type.

The third topic of the paper will be an implementation of these general concepts under the specific conditions of the molecular bridge model. If the bridge is non-interacting, it can be solved explicitly in full and consequently serve to elucidate the following general features:

- The initial transient dynamics as the reaction to the sudden or adiabatic changes in the system: This initial dynamics is described by the full NGF equations of motion with the finite time initial conditions explicitly included. To demonstrate this dynamics we calculate the response of molecular bridge system to changes of the boundary conditions created by switching on/off either of the leads, which results into uncorrelated or correlated initial conditions, and their subsequent influence on the dynamics of many body systems.

- Asymptotics of the bridge evolution at late times as described by GME: We will investigate the ways outlined above of a description of the dynamics of the bridge by a GME after the very initial stage of the transient. At this point we will discuss and compare various possibilities how to reconstruct correlation function from the single particle ”distribution” and their effect of the resulting form of the GME. The simplicity of the molecular bridge model enables us to discuss important details of this reduction; we will derive GME for the molecular bridge model explicitly and discuss their range of validity.

- From GME to a Master equation (ME): We will further simplify the GME to describe the final state of the evolution by ME. This reduction will be performed on three levels of sophistication. The lowest level (memory effect neglected, weak scattering) produces the standard Pauli Master equation, while the other two levels with an increasingly broad validity range lead to effective Master Equations with renormalized coefficients.

The non-equilibrium behavior of a system is determined within the NGF approach by a specific model of the NE self-energy components. There are several general properties of the self-energy which must be obeyed, like the spectral identity, and some other which we assume and which have to be verified in each specific instance. These include, in particular, two time scales, the fast one which relies on the existence of a pair of related characteristic times, the quasiparticle formation time and the collision duration time, and defines the shortest time structure, and the relaxation times related to the transport in the kinetic stage and to the quasi-particle decay. On the formal side, we keep the universality of the equations by avoiding any specific representation, so that the Green’s functions and related quantities appear mostly as having only time arguments explicit. Otherwise, they should be viewed as operators acting on one-particle states.

The paper is organized as follows: After this Introduction, the molecular bridge model and related NGF are introduced in Sec. II. In the following Sec. III. the NGF with a finite initial time are defined on the Schwinger-Keldysh contour, the concept of initial states as states reached by physical process is discussed, Dyson equations for NGF with initial conditions are introduced and
given in our notation for the self-energies including the singular parts of $\Sigma^\infty$. Corresponding selfenergies are then formulated with the time partitioning method. The NGF are introduced and Dyson equations are formulated for the molecular bridge model in Sec. IV. In Sec. V the dynamics of the molecular bridge in the first stage of its evolution, when initial conditions play an important role, is discussed. Based on the observation that some steady state could develop in later stages of the evolution, the following Sec. VI is devoted to the simplified single time asymptotic transport equations: it deals with the problem of quantum transport equations is addressed from a general point of view. First, the precursor quantum transport equation including the initial correlations is given and the Bogolyubov conjecture about a fast decay of initial correlations is presented. The precursor equation is then converted into a true transport equation of the GME type with the aid of the so-called causal ansatzes, a representative example of which is the Generalized Kadanoff-Baym Ansatz. The GME from this Section are then specified for the molecular bridge model in Sec. V. The results of this Section are then used for further simplification of asymptotic single time transport equations of the GME type to Markovian Master equations in Sec. VII. The specific choice of various parts of the Hamiltonian we have the following explicit expressions:

$$H'(t) = \alpha_L(t)V_L + \alpha_R(t)V_R$$  \hspace{1cm} (2)

The specific choice of various parts of the Hamiltonian is as follows. The three parts of the structure, taken as separate, have no time dependence. The leads, labeled by $L$ and $R$, couple only to the bridge $B$, but not one to another. The coupling is specified by time independent operator amplitudes $V_{L,R}$ coupling the leads to the central island orbital. All time dependence in the model is concentrated into two scalar functions $\alpha_L,\alpha_R(t)$. These functions may be arbitrary, but to describe the switching processes, the useful choice is to let them to jump suddenly between 0 and 1, describing a disconnected and connected junction respectively.

We further assume a single-level bridge. For all components of the Hamiltonian we have the following explicit expressions:

$$H_{\alpha_L} = \sum_{\ell} |\ell\rangle \varepsilon_{\ell}(\ell), \quad \varepsilon_{\ell} = \tilde{\varepsilon}_{\ell} + \Delta_L,$$

$$H_{\alpha_R} = \sum_{r} |r\rangle \varepsilon_{r}(r), \quad \varepsilon_{r} = \tilde{\varepsilon}_{r} + \Delta_R,$$

$$H_{\alpha_{ib}} = \langle|b\rangle \varepsilon_{ib}(b)\rangle$$  \hspace{1cm} (3)

Here, $|b\rangle$ is the single electron orbital at the island, the decay states $\{|a\rangle\}$ are further divided between two complementing orthogonal subspaces of the two leads: $\{|a\rangle\} = \{|\ell\rangle\} \oplus \{|r\rangle\}$, $\tilde{\varepsilon}_{\ell}$ and $\tilde{\varepsilon}_{r}$ are the 'bare' one-electron level in unbiased leads and the level shifts $\Delta_L, \Delta_R$, correspond to the independently floating grounding of both leads considered as extended electron reservoirs.

Finally, expressions

$$V_L = \sum_{\ell} |\ell\rangle v_{\ell}(b) + H. \ c.$$

$$V_R = \sum_{r} |r\rangle v_{r}(b) + H. \ c.$$  \hspace{1cm} (4)

introduce the coupling constants $v_{\ell}$ and $v_{r}$. It may be said that the detailed structure of the leads is responsible for the behavior of the system, because much of its physics is contained in the band energies $\tilde{\varepsilon}_{\ell}$ and $\tilde{\varepsilon}_{r}$, and in the coupling constants $v_{\ell}$ and $v_{r}$. In particular, if these functions are flat around the resonant energy $\varepsilon_h$, the so-called wide band limit (WBL) is achieved. We will be more interested in situations for which the bands are not "wide", because deviations from WBL give rise to specific behavior of the system at very early stages of a transient.

Due to the links of the central island to the leads, physics of this simple model well simulate behavior of the many body systems (for details see the discussion in [6]) and so, it is reasonable to treat the nonequilibrium dynamics of this model by the NGF technique, which is natural for the description of many body systems.

II. MOLECULAR BRIDGE MODEL

The prototype model of a nanoscopic structure is a single level molecular bridge between two current leads:

```
LEFT LEAD [橋] BRIDGE ISLAND [橋] RIGHT LEAD
```

The two leads are connected to the central island each by its own independent tunneling junction, left and right. This model has been studied extensively, as reviewed in [39]. The model sophistication has been gradually improved. For a fully systematic account of many-electron interactions based on the modern time-dependent density functional theory see [40]). For inclusion of phonon reservoirs see e.g. [41]. Less attention has been paid to the fast transient currents [6, 32, 42]. Most of the work on the molecular bridge model, including ours, has been based quite naturally on the use of non-equilibrium Green’s functions (NGF). Two fundamental references are [43] for stationary currents and, in particular, Ref. [44] for time-dependent phenomena.

The one-electron Hamiltonian for the bridge model is taken in its simplest possible form [6], specializing further the Hamiltonian in [44]:

$$H(t) = H_0 + H'(t)$$

$$H_0 = H_{\alpha_L} + H_{\alpha_R} + H_{\alpha_{ib}}$$  \hspace{1cm} (1)

The dynamics of the system is defined by the full Hamiltonian $H + U(t)$ consisting of the system Hamiltonian

A. NGF - basic definitions

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Here, $|b\rangle$ is the single electron orbital at the island, the decay states $\{|a\rangle\}$ are further divided between two complementing orthogonal subspaces of the two leads: $\{|a\rangle\} = \{|\ell\rangle\} \oplus \{|r\rangle\}$, $\tilde{\varepsilon}_{\ell}$ and $\tilde{\varepsilon}_{r}$ are the 'bare' one-electron level in unbiased leads and the level shifts $\Delta_L, \Delta_R$, correspond to the independently floating grounding of both leads considered as extended electron reservoirs.

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Due to the links of the central island to the leads, physics of this simple model well simulate behavior of the many body systems (for details see the discussion in [6]) and so, it is reasonable to treat the nonequilibrium dynamics of this model by the NGF technique, which is natural for the description of many body systems.
A dynamical process is described by the statistical operator \( \hat{P}(t) \) which is fully specified by its initial many-body state \( \hat{p}_i \) at \( t = t_i \) (an arbitrary equilibrium or out-of-equilibrium state).

The Non-equilibrium Green’s Function is defined in the usual manner \([1, 2, 18, 19]\),

\[
G(1, 2) = -i \text{Tr} \left( \hat{p}_i \hat{T}_c \{ \psi(t_1) \psi^\dagger(2|t_1) \} \right)
\]  

(5)

with the Heisenberg field operators \( \psi, \psi^\dagger \) anchored at \( t_1 \) and the time-ordering operator \( \hat{T}_c \) acting along the closed time path \( \mathcal{C} \) extending from \( t_1 \) to \( +\infty \) and back.

We represent the contour-ordered NGF, following Keldysh \([12, 14, 16]\), by a \( 2 \times 2 \) matrix GF of real time, employing the Langreth-Wilkins matrix \([1, 2, 5, 13, 18, 45]\) having three components, the less-correlation function \( G^c \) and two (equivalent) propagators, \( G^A(1, 2) = [G^R(2, 1)]^\dagger \).

**B. Initial conditions**

In general, the initial condition for the NGF is given by an arbitrary initial many body state \( \hat{p}_i \) at \( t = t_i \). Its time \( t_i \) determines the beginning of the Schwinger trajectory on which the NGF is defined, see Fig. 1a.

\[
\begin{align*}
\mathcal{C}_1 & \quad t_1 \equiv t_v \\
\mathcal{C} & \quad t_v \end{align*}
\]

Fig. 1: The NGF time contour and its extensions.

The process under study evolves along the Schwinger-Keldysh trajectory starting and ending at \( t_i \).

a. Schwinger trajectory \( \mathcal{C}_1 \) of the process under study. The initial time \( t_i \) is put to \( t_v \), selected as the onset of the relevant non-equilibrium process.

b. Keldysh trajectory \( \mathcal{C}_-\infty \) for a “host” process starting at \( t_i \rightarrow -\infty \). The \( t_v \) point marks again the onset of non-equilibrium.

The natural question is how to reach such an initial state. To answer this question, it is reasonable to assume that it is prepared by a previous physical process. To this end, we can start to evolve the measured system from an equilibrium non-perturbed state at an initial time \( t_i \rightarrow -\infty \) by a slow, adiabatic, switching on of the interactions, as is well known from the perturbation theory of many-body systems - this corresponds to an extension of the Schwinger trajectory to \( t_i \rightarrow -\infty \), see the Fig. 1b. The original trajectory is a part of the extended trajectory and its starting point is renamed to \( t_v \).

In this case, \( t_v \) coincides with \( t_v \), the end point of a well-defined physical process, the Keldysh prelude, by which we gain a dressed (correlated) equilibrium state right at the onset of the real non-equilibrium dynamics. We will call the extended process the switch-on state, and the corresponding trajectory, on which the NGF is defined, the Keldysh trajectory, see Fig. 1b.

Fig. 1 b is redrawn in a slightly different format in Fig. 2a. The whole switch-on process consists of the Keldysh prelude and of the relevant process starting at \( t_v \). We can now generalize this Keldysh method to the case of the correlated initial state out of equilibrium, provided it has been obtained as an outcome of the previous evolution of the system. This can be achieved, if the Keldysh prelude serving as a reference is followed by a preparatory non-equilibrium process lasting from \( t_v \) to \( t_{\text{end}} \). The state reached by the system at \( t_{\text{end}} \) then serves as the initial state for the ensuing evolution of the system during the measurement period. In other words, from the whole host process, we single out a relevant sub-process whose initial time \( t_v \) coincides now with \( t_{\text{end}} \). This division of the whole switch-on process is defined on purely operational grounds, and its usefulness depends on the existence of a suitable way of constructing the NGF within the measurement stage. It is thus productive to think of the entire switch-on state as of a host process consisting of the three stages, and of the measurement stage as of the embedded sub-process. This sequential division of the host process is illustrated in Fig. 2b.

It is clear that the initial states in our approach form a restricted set of those states which can be achieved by physical means, that is by a dynamical distortion of an equilibrium state. In other words, we are limited to the natural initial conditions, while artificial initial states, say overcorrelated, are excluded. This distinguishes the present approach from other techniques admitting also initial states which cannot be generated by natural means.

**C. Dyson equation with initial conditions**

Let us return to the basic Schwinger contour starting at \( t_i = t_v \), see Fig. 1a. As already mentioned in the Introduction there are several formulations of the perturbation theory for the NGF with the initial conditions \([1, 5-7, 15, 25]\), but all of them provide us the same structure of Dyson equations, which will be now described.

It is convenient to write the Dyson equation first by components. For the propagators, it will have the form

\[
G^{R,A} = G^{R,A}_0 + G^{R,A}_0 \Sigma^{R,A} G^{R,A}, \text{ etc.}
\]  

(6)

in which the self-energies are ”regular” two-time functions. The mean-field ”singular” parts of the self-energy are included into the free propagators. The less-
The initial condition at part. The outcome of the preparation history is distilled into is the period of preparation followed by the relevant process 

b. The case when only a part of the process is relevant, study specifically; light stretches symbolize the switching processes we want to 

The "interactions" simulated by the coupling to the leads are 

The four terms have a varying degree of singularity at 

Note that the integrations in (7) start at 

Note the change in the name of the lower integration limit. Now it 

also with the decomposition (8) and write it in an explicit form:

and are thus equivalent to single-time continuous functions 

The last term,

represents the uncorrelated part of the initial conditions. This is the only part of \( \Xi^< \) which enters the free particle correlation function

D. Time partitioning

In this section, we introduce the perturbation scheme which enables us to write closed equations for the NGF describing time evolution of the system during the non-equilibrium process with inclusion of the initial conditions at \( t_P \), as expressed in terms of the history of the system between \( t_1 \to -\infty \) and \( t_P \). This is sketched in Fig. 1b. We aim at applying this approach to the initial conditions for the non-equilibrium dynamics of the molecular bridge model.

This perturbation scheme is based on the time partitioning method, see [5–7]. We will not go to details of its derivation here, see [7]. To present the partitioned form of \( G^< \), it is convenient to combine the Dyson equation (7) with the decomposition (8) and write it in an explicit form:

Note the change in the name of the lower integration limit. Now it \( t_P \), while \( t_1 \) has been shifted to the remote past at the onset of the Keldysh prelude. The whole host (switch-on) process is partitioned into the past prior to \( t_P \) and into the future after \( t_P \). This partitioning is reflected in the form of the components of the less self-
energy entering Eq. (12),

\[ A^<(t, u) = i \int_{t_1}^{t_2} dt' \{ G^R(\Sigma^c + G^< \Sigma^A) \} \]
\[ A^<(v, t) = -i \int_{t_1}^{t_2} dt' \{ \Sigma^c G^A + \Sigma^R G^< \} \]
\[ \Sigma^c(v, u) = \Sigma(v, u) \]
\[ \int_{t_1}^{t_2} dt' \{ \Sigma^R G^R \Sigma^c + \Sigma^R G^< \Sigma^A + \Sigma^c G^A \Sigma^A \} \]
\[ \Sigma^R G^R \Sigma^c \to \Sigma^R(u, t) G^R(t, t') \Sigma^c(t', v), \text{ etc.} \] (13)

By these relations, the self-energy of the relevant process is expressed by integrals involving time blocks of the Green’s functions and the self-energies of the host process. The external arguments \( u, v \) refer to the process in the future and are always greater than \( t_\nu \), while the integration variables, denoted by \( \tilde{t}, \tilde{t'} \) for clarity, belong entirely to the past and are less than \( t_\nu \). The propagation takes place entirely in the past. The history and the future are interconnected by the off-diagonal blocks of the self-energies. The singular components of the self-energy have no analogue in the host process, while the regular term \( \Sigma^c \) has two parts

\[ \Sigma^c = \Sigma^c(t, t') + \tilde{\Sigma}^c(t, t') \] (14)

The first term comes from the host process without change (where it would enter the Dyson equation in the usual form, \( G^c = G^R \Sigma^c G^A \)). It is supplemented by the second term, \( \tilde{\Sigma}^c(t, t') \), which takes the finite time initial condition into account.

1. Properties of host self-energies and initial conditions

To identify the functions \( A^c, A^<_c, \Sigma^c \) as describing finite time initial conditions, which are expected to decay after some characteristic time \( \tau_c \), it is necessary to make an assumption about the time behavior of the host self-energies \( \Sigma^R, \Sigma^c \), which determine behavior of these functions.

For many systems it has been found that self-energies are basically negligible, if their two time arguments are more apart from each other than some characteristic time \( \tau^* \). This is in correspondence with the Bogolyubov principle, which postulates that all temporal correlations in the system (including, of course, the initial correlations) decay within a period with a characteristic "Bogolyubov" time \( \tau_\nu \). This time typically turns out to be comparable with the \( \tau^* \) time. The Bogolyubov principle is thus translated into the assumption about the behavior of the system expressed in terms of self-energies: All components of the host self-energy should be concentrated to a strip

\[ |t - t'| < \mathcal{O}(\tau^*) \quad \tau^* = \mathcal{O}(\tau_c, \tau_\nu) \] (15)

The two times appearing at the r.h.s. are: \( \tau_c \), often called the collision duration time in transport theory, relevant for \( \Sigma^c(t, t') \), and \( \tau_\nu \), usually called the quasiparticle formation time, characteristic for \( \Sigma^R, \Sigma^A(t, t') \).

With the above assumption about the restricted time range of self-energies, all integrals in (13) have a correspondingly restricted integration region given by the condition (15). Neither the self-energies nor the Green’s functions have to be known deeper into the past than to \( t_\nu - \mathcal{O}(\tau_\nu, \tau_c) \). This is a much reduced information actually required, rather than the knowledge of the preparation process over the whole endlessly extended past. Consequently, the self-energy corrections (13) become a manageable and convenient means for incorporating the correlated initial conditions into the finite time NGF formalism. We have thus achieved a practicable implementation of the diachronous approach to the initial condition problem: the initial condition is captured through the history of the system as reflected in single particle correlation functions. The actually required depth of the memory into the past is finite, comparable with the decay time of the correlations into the future. This diachronous picture should be compared with the synchronous alternative of describing the initial conditions by higher order correlations, all at the initial time \( t_\nu \).

Looking into the future, we can see that for the same reason, all integral terms in (13) will be vanishingly small for their external arguments greater than \( t_\nu + \mathcal{O}(\tau_\nu, \tau_c) \). Beyond this time characterizing the decay of correlations in the system, only \( \Sigma^c \) of the host process will survive. By this, we may conclude that the initial transient in the Green’s function \( G^c \), Eq. (12), will vane out over a time equal to the correlations decay time plus the propagator relaxation time beyond \( t_\nu \). After that, the common form \( G^c = G^R \Sigma^c G^A \) will prevail.

2. Notes to the time partitioning method

So far, we have considered the time splitting of the whole host process at \( t_\nu \), as illustrated by Fig. 2a. It is clear that the time partitioning is universal and does not depend on any assumptions, like equilibrium, about the "past". We may thus shift the splitting time to any other convenient instant. In particular, it will be advantageous to choose \( t_M \), as illustrated in Fig. 2b. In other words, the future will coincide with the relevant period of measurement on the system. The preparatory stage will then be probed by the partitioning integrals again not deeper than by about \( \tau^* \), but with respect to \( t_M \). It is enough to wait with \( t_M \) for a time \( \sim 2\tau^* \) past \( t_\nu \) and this dividing time between the Keldysh prelude and the active preparation will not be included. For a shorter time separation of both instants, an interference of both transients may play role.
IV. NGF FOR THE BRIDGE MODEL

In this chapter, before we use the partitioning in time method to calculate and discuss the non-equilibrium dynamics of the molecular bridge in detail, we will explore the simplicity of the molecular bridge model to shed some light on the standard NGF approach and the role of initial conditions. Before proceeding with the standard construction of the NGF for the dynamics of the molecular island, we will first write the NGF for the whole system directly, making use of the circumstance that for non-interacting electrons, the NGF of the whole nanoscopic structure can be quite generally reduced to an unitary dynamics within the space of one-electron states. Then we will create the NGF for the island by the projection on the island state. The dynamics of the island will be then determined by the self-energy, which is created by this projection procedure (partitioning in the Hilbert space).

This dynamics thus parallels the behavior known from many-body systems, where interactions are described by corresponding self-energies.

To indicate the global nature of the one-electron quantities for the whole system, we will use calligraphic letters:

\begin{align}
G^R(t,t') &= -i \mathcal{S}(t,t') \theta(t-t'), \\
G^A(t,t') &= i \mathcal{S}(t,t') \theta(t'-t) \\
G^<(t,t') &= i \mathcal{S}(t,t_i) \mathcal{g}_i \mathcal{S}(t_i,t') 
\end{align}

Here, \( \mathcal{S} \) denotes the one-electron evolution operator. Density matrix

\begin{align}
\mathcal{g}(t) &= \mathcal{S}(t,t_i) \mathcal{g}_i \mathcal{S}^\dagger(t,t_i) \\
i \partial_t \mathcal{S}(t,t') &= \mathcal{H}(t) \mathcal{S}(t,t'), \quad \mathcal{S}(t,t) = I_{op}
\end{align}

represents a formal solution of the time development of the bridge specified by the one-electron Hamiltonian \( \mathcal{H}(t) \), and by the initial one-electron density matrix \( \mathcal{g}_i \) (initial OEDM at \( t_i \)).

Introducing the projection operators

\begin{align}
\mathcal{P} &= |b\rangle \langle b| \\
\mathcal{Q} &= \sum_a |a\rangle \langle a| \\
I_{op} &= \mathcal{P} + \mathcal{Q}, \quad \mathcal{P} \mathcal{Q} = 0
\end{align}

we can define the "true" NGF for the bridge by the \( \mathcal{P} \)-projection of the GF defined in Eqs. (16),(17)

\begin{align}
\mathcal{G}^b \rightarrow \mathcal{P} \mathcal{G}^b \mathcal{P} \\
= |b\rangle \langle b| \mathcal{G}^b |b\rangle \langle b| \equiv \mathcal{P} \mathcal{G}^b \\
\mathcal{G}^b = |b\rangle \langle b| \mathcal{g}_b, \quad z = R, A, <
\end{align}

Now, to follow the common procedure known from the many-body systems, we have to write equation of motion for the NGF of the island, i.e. we have to formulate the perturbation scheme and the corresponding Dyson equations. We will start with propagators.

To obtain the Dyson equations for the propagators \( \mathcal{G}^{R,A} \), we start from the purely unitary Dyson equations for their global counterparts

\begin{align}
\mathcal{G}^{R,A} &= \mathcal{G}_0^{R,A} + \mathcal{G}_0^{R,A} \mathcal{H}' \mathcal{G}_0^{R,A} \\
\mathcal{G}_0^{R,A} &= \mathcal{G}_0 + \mathcal{G}_0^{R,A} \mathcal{H}' \mathcal{G}_0^{R,A}
\end{align}

Here, the unperturbed NGF are associated with \( \mathcal{H}_0 \). The time integrations extend over the natural limits, like

\[ \mathcal{G}^R = \cdots + \mathcal{G}_0^R \mathcal{H}' \mathcal{G}^R \rightarrow \mathcal{G}^R(t,t') = \cdots + \int \mathcal{H} \mathcal{G}(t) \mathcal{H}'(\tau) \mathcal{G}^R(\tau,t') \]

The \( \mathcal{G}_0^{R,A} \) GF are block diagonal together with \( \mathcal{H}_0 \), while the coupling \( \mathcal{H}' \) is block off-diagonal. This permits the standard partitioning procedure [47] leading at the end to four genuine Dyson equations as counterparts to Eqs. (24). For example, the first of these equations reads

\begin{align}
\mathcal{G}^R &= \mathcal{G}_0^R + \mathcal{G}_0^{R} \mathcal{R} \mathcal{G}^R \\
\mathcal{R} &= \{ b \} \mathcal{H} \mathcal{G}_0^R \mathcal{Q} \mathcal{Q} \mathcal{H}' \{ b \}
\end{align}

The self-energy \( \mathcal{R} \) has a clear interpretation: an electron in the bridge state is virtually scattered to the lead states and back.

We can interpret this self-energy as the description of a fictive interaction which is presented in the model, so we can clearly see that the existence of this self-energy enables us to formulate the analogy between the behavior of the molecular bridge model and real many body systems: the transitions between the island state and the decays states of the leads are caused by the island-leads couplings, while the transitions between the single particle excitations (single quasiparticle states) and the decay (multi-particle) states in many body systems are caused by multi-particle interactions, for details see [6].

B. Dyson equation for \( G^< \)

The same elementary procedure permits to obtain explicit expressions for the self-energy \( \Xi^< \) entering the Dyson equation (7) for the particle correlation function \( G^< \). To derive expressions for these quantities, we first rewrite Eq. (17) for \( G^< \) using the definitions (16)

\begin{align}
G^<(t,t') &= i \mathcal{S}(t,t_i) \mathcal{g}_i \mathcal{S}(t_i,t') \\
G^<(t,t') &= i \mathcal{G}^<(t,t_i) \mathcal{g}_i \mathcal{G}^A(t_i,t')
\end{align}

Then we introduce \( \mathcal{P} + \mathcal{Q} = I_{op} \) (22) into the definition (23) of \( G^< \):

\begin{align}
G^<(t,t') &= \langle b| G^< (t,t') | b \rangle \\
&= i \langle b| \mathcal{G}^R(t,t_i) (\mathcal{P} + \mathcal{Q}) \mathcal{g}_i (\mathcal{P} + \mathcal{Q}) \mathcal{G}^A(t_i,t') | b \rangle
\end{align}
The four term structure of $\Sigma^< \!$ of (7) results immediately by multiplying through the $2 \times 2$ parentheses in Eq. (27) and introducing the equations (24) into each of the terms. The final result is

$$\begin{align*}
\rho_t &= \langle b | g_t | b \rangle \\
A^<(t, t') &= i \langle b | g_t QG_0^R(t, t') Q \mathcal{H}'(t') | b \rangle \\
A^\gamma(t, t') &= i \langle \mathcal{H}'(t) Q G_0^R(t, t') | b \rangle \\
\sum^<(t, t') &= i \langle b | \mathcal{H}'(t) Q G_0^R(t, t') Q \mathcal{H}'(t') | b \rangle
\end{align*}$$

(28)

Here, the two symmetrically formed $A^<$, $A^\gamma$ enter the "mixed" terms $\sum^<$, $\sum^\gamma$ of the Dyson equation (7). These both terms involve the off-diagonal components of the initial density matrix $g_t$ (commonly denoted as coherent terms) and vanish together with them. These off-diagonals of $g_t$ are a signature of "correlated initial conditions", just like the interactions are modeled by the off-diagonal $P - Q$ mixing. The two remaining terms, $\rho_t$ and $\sum^\gamma$, correspond roughly to the self-energy for uncorrelated initial conditions. They depend on the diagonal blocks of $g_t$ and do not vanish even if $g_t$ is block-diagonal.

Looking at the above expressions for the correlation function of the selfenergy we can see immediately two important features of the model. First, the present construction of the selfenergy witnesses the analogy between the dynamical behavior of non-interacting molecular bridge model and of the many body systems extended also to the statistical features now.

Second, regarding initial conditions, this approach is an instructive example of the synchronous method, in which all correlations at the initial time are included in full, that is exactly.

We can now compare this result gained by the synchronous method with the result obtained by the time partitioning which represents the diachronous treatment of the initial condition. As is shown in [6], in the case of the molecular bridge model, it is possible to transform these results onto each other explicitly. This provides a unique insight into the details of the equivalence between the diachronous approach coached in the field theoretic veil and the more transparent synchronous description of the model.

C. The host NGF

Our next aim is to explore the dynamics of the bridge starting from a well defined initial state, be it uncorrelated or correlated. As explained in Sec. III, to this end we first need the characteristics of the host process, specifically we have to construct its self-energies.

In the case of structures similar to the molecular bridge, such self-energies were introduced in the article of Jauho, Wingreen and Meir [44] for the first time, so we will call these functions the JWM self-energies.

The JWM results are easily reproduced within our approach based on the partitioning in Hilbert space; it is straightforward to obtain the JWM functions from the definitions (7), (8), (9), and the general solution (28). These equations are adjusted to the host (switch-on) process by simply shifting the initial time to infinity, $t_0 \equiv t_{-\infty} \rightarrow -\infty$. The usual assumption of a time-independent unperturbed Hamiltonian is generalized somewhat to

$$P_{\mathcal{E} \rightarrow -\infty} Q = P_{\mathcal{E} \rightarrow -\infty} P = 0$$

(29)

$$[\mathcal{E}_{\mathcal{E} \rightarrow -\infty} Q, \mathcal{H}_f(t)] = 0$$

(30)

The initial state is uncorrelated by the first line, but an additional enhanced condition (30) is needed to bring $G^<$ to the JWM form

$$G^< = G^R \Sigma^< G^A$$

$$\Sigma^R = \langle b | \mathcal{H} Q G_0^R | b \rangle$$

$$\Sigma^A = \langle b | \mathcal{H} Q G_0^A | b \rangle$$

$$\Sigma^< = \langle b | \mathcal{H} Q G_0^< | b \rangle$$

(31)

where

$$G^<_B(t, t') = i G_0^R(t, t_{-\infty}) e^{-i t_{-\infty} \mathcal{H}^B} G_0^A(t_{-\infty}, t)$$

(32)

The remarkable feature of the JWM self-energy is that, contrary to the appearance, it contains in fact only the external times, as follows by Eqs. (29) and (30). The expressions (31) can then be rewritten in terms of the spectral density $A_0(t, t') = \{G_0^R(t, t') - G_0^A(t, t')\}$ with the result:

$$\Sigma^R(t, t') = -i \langle b | \mathcal{H}'(t) Q A_0(t, t') Q \mathcal{H}'(t') | b \rangle \delta((t - t'))$$

$$\Sigma^A(t, t') = +i \langle b | \mathcal{H}'(t) Q A_0(t, t') Q \mathcal{H}'(t') | b \rangle \delta(t' - t)$$

$$\Sigma^<(t, t') = i \langle b | \mathcal{H}'(t) Q A_0(t, t') Q \mathcal{H}'(t') | b \rangle$$

(33)

Using the decomposition introduced in Eqs. (1) and (2), an explicit integral representation for the above self-energies is obtained:

$$\Sigma^\gamma(t, t') = \Sigma^\gamma_0(t, t') + \Sigma^\gamma_\delta(t, t')$$

$$\Sigma^\gamma(t, t') = \alpha_\gamma(t) \sigma^\gamma_\delta(\tau) \alpha_\gamma(t')$$

$$\sigma^R, A(\tau) = \mp i \int \frac{dE}{2\pi} \Delta_Y(E) e^{+iE\tau} \delta(\pm \tau)$$

$$\sigma^<(\tau) = i \int \frac{dE}{2\pi} \Delta_Y(E) f_Y(E) e^{-iE\tau}$$

$$\tau = t - t', \quad \tau + t = R, \quad A, \quad Y = L, \quad R$$

(34)

In this way, all components of the JWM self-energy for the bridge model have the form reflecting the presence of two leads. Each fully connected junction is thus associated with a spectral function $\Delta_Y$, which, together with the quasi-equilibrium Fermi function $f_Y$, specifies the $Y$ self-energy triplet.
V. QUANTUM TRANSPORT AND INITIAL CONDITIONS

The question we will deal with in this section is, how a system, in this case the molecular bridge, will react to nonequilibrium conditions in dependence on its intrinsic times.

In principle, any time-dependent process at the bridge is solved by the set (34) of expressions for the self-energies. Our goal, however, is to explore the dynamics of the bridge model during the measurement process starting from a correlated initial condition at a finite time \( t_{\text{M}} \). This process will be viewed, following the sketch in Fig. 2b, as embedded into the complete Keldysh "host" process as discussed in previous sections, see also [6].

This embedding does not affect the propagators, but the Dyson equation (12) for the island less function becomes more involved, because it contains terms describing the initial correlations. Technically, the embedding means to construct these corrections from the self-energies of the host process (34) with the help of the time partitioning formulas (13).

We have now formalism ready to calculate the response and transport characteristics of the molecular bridge to the changes in its boundary conditions: switching the leads on and off, and we can calculate the response to either the uncorrelated or the correlated no-equilibrium initial conditions. Before we show some results, it will be profitable to discuss in a more detail the physics involved in these calculations.

A. Models used for transport calculations

We will investigate the molecular bridge model in which the left and right leads have a different bias with respect to the island level, but both leads represent equilibrium reservoirs having their Fermi Dirac distribution (in general, they might have different not only their chemical potential, but also the temperatures). The spectral functions \( \Delta Y \) float together with the respective chemical potentials, whose difference is \( \Delta V \), see Fig. 3.

The molecular island has only one level, consequently no own dynamics; all dynamical behavior is controlled by the junctions, and depends on the properties of the reservoirs. This dynamics and the intrinsic times of the model are thus contained in the host process retarded JWM selfenergy specified in turn by , Eq. (34). These spectral functions is in fact physically very close to the adsorption function of Grimley or Newns [46] and may have a pronounced spectral structure which is transferred onto the short time behavior of the self-energy [6]. When we model the true spectral function by a sharp peak, see Fig. 3, its width is inversely proportional to a characteristic time which we can identify as the quasiparticle formation time \( \tau_Q \) entering Eq. (15).

The magnitude of \( \tau_Q \) influences the dynamical behavior of the system significantly. In the limit of small \( \tau^* \), the propagators have a negligible formation period and all of the short time transient behavior depends on \( \tau_c \) given by the relative position and the width of the statistical distributions entering \( \Sigma^< \) in (34). This limit corresponds to a flat spectral function and is commonly known as the Wide band limit (WBL), see Fig. 4. Calculations of the transport properties of the molecular bridge models in the literature are mostly done using this convenient limit. The opposite case, the narrow band limit (NBL) sketched in Fig. 3 brings about a more complex short time dynamical behavior, reflecting the interplay of both energy dependent structures, the statistical distributions and the spectral functions. Most of our calculations will be performed in this limit.

B. Response to switching the junctions on and off

For actual calculations, the two metallic leads were taken as identical. The spectral function \( \Delta_Y(E) \) combined a broad background with a resonant structure (FWHM 0.3 eV). This, together with the step-like distribution function \( f_Y(E) \) gave rise to \( \tau_c \) in the 10 fs range. The sharp switching, assumed in our model, may thus simulate events with femtosecond switching times. The leads had a constant bias \( \pm 0.5 eV \) with respect to the island level, such that the charge flowed from the \( L \) lead to the \( R \) one on the whole. During the "prelude" up to the initial time \( t_0 = 0 \) of the transient, the level was empty and disconnected from both leads. For \( t > t_0 \), the currents through both junctions were calculated using the JWM selfenergies (34), and the time partitioning technique discussed in Sec.III, for details see [5–7].
FIG. 4: Electronic structure of the bridge model – wide band limit. The broad spectral density is shown as the light gray profile and partly overlapped by the lead FD distribution function.

1. Island suddenly connected to the leads: uncorrelated initial condition

As the first case, we show the transient induced by switching both junctions on at \( t_1 = 0 \). This is the simple case of uncorrelated initial conditions studied previously in the wide band limit in [42]. Fig. 5, left panel, shows transient oscillations of the currents \( J_L, J_R \) through both junctions, followed by balanced steady currents after elapsing about 20 fs. The occupancy rate of the island level, shown in the right panel, first oscillates and saturates to zero correspondingly. A deeper insight is provided by Fig. 6 showing \( G^< \) as a double time function. The crest line of the plot along the diagonal \( t = t' \) is the occupation number \( \rho(t) = iG^<(t,t) \). It exhibits three stages: initial oscillatory transient, gradual saturation controlled by the transport relaxation time, and finally the steady state. The time derivative of this line reproduces the occupancy rate of Fig. 5. Perpendicular to the time diagonal, the plot exhibits slowly evanescent coherent oscillations. Along the time diagonal, they persist, however.

2. Multiple switching: correlated initial condition

The next simplest case is specified by the sequence of two switching events: \( \diamond t \geq t_0 = 0 \) fs : \( L \) on, \( R \) off, \( \diamond t \geq t_1 = 40 \) fs : \( L \) on, \( R \) on.

In Fig. 7, the left panel, the currents through both junctions are shown again. As before, each of the two switchings gives rise to a transient – the transitory current oscillations, which die out gradually. The first event opens only the left junction, only one \( J_L \) current is non-zero and the level gradually fills up. The second switching is a novel feature, as it starts from a correlated initial condition, generated by the "history", that is the previous evolution initiated by the first switching event. The principal effect is the opening of the second, \( R \), junction which starts to abduct the electrons from the island. The sign of the initial oscillation is reverted, therefore. The \( R \) junction “overreacts” at first and this is compensated later by the continued transient. It should be noted that the \( L \) junction invariably responds, with a slight delay, to the state of the \( R \) junction. Because the two junctions act differently having a mutually opposite bias, the \( L \) junction contributes to the compensation of the initial transient at the \( R \) junction. This is neatly demonstrated at the total occupancy rate in the right panel of Fig. 7.
3. Summary of results

We can observe at least two clear stages in the evolution: first, the "wild" period when values of the observables fluctuate and are strongly dependent on the initial conditions. This stage is followed by a milder time development. We can clearly see the time asymptotics in all figures shown: some time after the last changes of the boundary conditions, time evolution of the observables slows down and even tends to stabilize. The natural question emerges, if these stages of evolution would be possible to describe by a time asymptotic evolution, i.e. to use the GME or even the ME for the description of these evolution stages of the molecular bridge dynamics. To explore this possibility, we start by considering the assumptions under which the NGF double-time transport equations can be approximated by a single time transport equation of the GME type in general.

VI. LONG TIME ASYMPTOTIC: TOWARDS QUANTUM TRANSPORT EQUATIONS

The aim of this chapter is to discuss conditions under which it is possible to simplify the structure of NGF to obtain a single time quantum transport equation by a well defined reduction procedure.

A. Precursor quantum transport equation

The well known starting point on the way from the double time NGF to single time transport equations is the so-called a Generalized Kadanoff-Baym Equation. It is a differential equation obtained from Eq. (7), and Eq. (6) written as \([G^{R,A}]^{-1} = [G_0^{R,A}]^{-1} - \Sigma^{R,A}\).

\[
G_0^{-1} G^< - G^< G_0^{-1} = \Sigma^R G^< - G^< \Sigma^A - G^R \Sigma^< + \Sigma^< G^A \tag{35}
\]

This (exact) equation has already a structure closely related to transport equations. The four terms on the r.h.s. represent the generalized collision terms. It still has the double time structure, and, in contrast to common transport equations, it also incorporates the initial conditions through the correlation function of the self-energy \(\Sigma^<\). Using Eq. (7) and splitting the self-energy \(\Sigma^<\) into the self-energy \(\Sigma^<\) of the host process (for the bridge model this is just the JWM selfenergy) and the remainder \(\Theta^<\) including all contributions from the initial correlations:

\[
\Theta^< = \Sigma_0^< + \Sigma_1^< + \Sigma_2^< + (\Sigma_0^< - \Sigma^<),
\]

we can rewrite Eq. (35) into the following form:

\[
G_0^{-1} G^< - G^< G_0^{-1} = \Sigma^R G^< - G^< \Sigma^A + \Sigma^< G^A - G^R \Sigma^< + \Theta^< \Sigma^A - G^R \Theta^<. \tag{36}
\]

Next, we go to the limit \(t = t'\) in order to obtain a single time transport equation – the precursor quantum transport equation. At equal external times, \(t = t'\), the one particle density matrix \(\rho\) is given by

\[
G^<(t, t) = i\rho(t) \tag{37}
\]

Using then the explicit expression for \(G_0^{-1}\)

\[
G_0^{-1}(t, t') = \{i\partial_t - H_0\} \delta(t - t') = \{-i\partial_\tau - H_0\} \delta(t - t') \tag{38}
\]

the l.h.s. of the last equation then is transformed to an unrenormalized drift of the one-particle density matrix \(\rho\):

\[
\text{l.h.s. of (35)} \overset{t = t'}{=} \frac{\partial \rho}{\partial t} + i [H_0, \rho]_-	ag{39}
\]

This already has the form of a QTE, in which \(H_0(t)\) is the mean field one-particle Hamiltonian, and the whole equation Eq. (36) becomes the precursor of quantum transport equations:

\[
\frac{\partial \rho}{\partial t} + i [H_0, \rho]_- = - (\Sigma^R G^< - G^< \Sigma^A) + G^R \Sigma^< - \Sigma^< G^A + \Theta^<_0 G^A - G^R \Theta^<_0. \tag{40}
\]

The "generalized collision" terms on the r.h.s. still involve double time less quantities. The related integrals extend only to the past because of the presence of the propagator factors.

We want to use this equation for times of steady external conditions. More precisely, for times which are sufficiently separated from the time \(t_{\text{last}}\) of the last perturbation of the system. To understand how this can simplify the precursor equation, we have to return to the time structure of the self-energies.

B. Assumptions about the time structure of the self-energy

Again, as in Sec.III, the assumption we make is that there exists a time \(\tau^*\), such that

\[
\Sigma^>(t, t') \approx 0 \text{ for } |t - t'| > \tau^* \tag{41}
\]

This condition should not be taken too literally, as, e.g., the band edges lead to weak power law tails. It captures in a simple manner the essential feature of a typical self-energy, to be concentrated along the time diagonal, however, and is in agreement with the notion of the decay of correlations, as explained above. We will use (41) for convenience and simplicity. The finite range of the IC corrections is just a corollary.

To see, how the condition (41) works, we first of all note that the last two generalized collision terms of Eq. (40), which take the initial conditions into account explicitly, become zero as the running time \(t\) exceeds \(t_{\text{last}} + 2\tau^*\).
This is in accord with the Bogolyubov conjecture. Thus, it is enough to wait for just $2\tau^*$ past $t_{\text{last}}$, and the precursor QTE is free of any IC terms:

$$\frac{\partial \rho}{\partial t} + i[H_0, \rho] = -(\Sigma^R \rho^< - \rho^< \Sigma^A) + (G^R \rho^< - \rho^< G^A)$$  \hspace{1cm} (42)

In the remaining collision terms, the time integration range is restricted by the condition (41) to a finite interval $(t - \tau^*, t)$. This is the memory depth of the system. For a typical term in Eq. (42), $\Sigma^R \rho^<$, this means

$$\{\Sigma^R \rho^<\}(t, t) = \int_{t-\infty}^{t} d\tau \Sigma^R(t, \tau) \rho^<(\tau, t)$$

$$\rightarrow \int_{t-\tau^*}^{t} d\tau \Sigma^R(t, \tau) \rho^<(\tau, t)$$  \hspace{1cm} (43)

For $t > t_{\text{last}} + 3\tau^*$, the self-energy in (43) extends already over the steady time span, and while the turbulent past may be reflected in the values of $\rho^<$, it will in no case show in the form of the EOM (42). And this is all the Bogolyubov conjecture says.

C. Reconstruction theorem

To achieve the quantum transport equation proper, another standard step is needed, in which all double-time less quantities entering the "generalized collision" terms of the precursor quantum equation (42) will be replaced by expressions involving only the single time distribution of particles $\rho$. In general, such expressions cannot be given explicitly, but there exist the so-called reconstruction equations with $\rho$ an input and $\rho^<$ as the output. This gives rise to the cycle [1, 2, 5]

$$\text{reconstruction eq.} = \text{precursor transport eq.}$$  \hspace{1cm} (44)

constituting the transport equation alternative for generating the full $\rho^<$ correlation function. The reconstruction equations are thus not stand-alone equations, but one part of a linked twin process whose other constitutive part is a transport equation obtained in the indicated solution cycle from the precursor transport equation: $\rho^<$ is substituted from the reconstruction equations, the transport equation is solved for the density matrix $\rho(t)$ and this in turn enters the reconstruction equation as an input.

D. GME

The $\{\text{RE := precursor TE}\}$ procedure, exact in principle, can be shortcutted by using the zeroth order approximation for the solution of the reconstruction equations. This simplifies the problem greatly and leads directly to the GME for the density matrix. This approximation is known under the name of the Generalized Kadanoff-Baym Ansatz (GKBA) [1, 5, 18, 19, 37, 38] or its variants.

We first summarize the general method of generating a quantum transport equation from the precursor transport equation standard for the Keldysh initial conditions at $t_0 \rightarrow t_1$ [1, 2, 18, 19]:

- The double-time $\rho^<(t, t')$ is replaced by an approximate ‘Ansatz’ expression involving only its time diagonal $\rho(t) = -i\rho^<(t, t)$ and propagators.
- This expression is also introduced into the self-energy, for which a self-consistent approximation specifying the self-energy in terms of the Green’s function is assumed.
- As a parallel input the knowledge of the propagator components of the Green’s function in necessary. In general, the components of the NGF are interconnected, but often this is of a lesser importance and the propagators can be found beforehand once for ever.

E. From an Ansatz to the GME

The resulting transport equation depends on the approximate replacement of $\rho^<$. The historically first one is the famous Kadanoff-Baym Ansatz [11, 18], schematically $G^< = \frac{i}{2} [\rho, G^A - G^R|^+]$. This choice was connected with the subsequent quasi-classical expansion made by Kadanoff and Baym. This is irrelevant for systems with discrete levels. We will concentrate on the so-called XKB Ansatzes, belonging to the general class of the causal Ansatzes, and exemplified by the original Generalized Kadanoff-Baym Ansatz – GKBA [18, 37, 38]

$$G^< = -G^R \rho + \rho G^A$$  \hspace{1cm} (45)

This ansatz is exact for independent particles governed by a one-particle Schrödinger equation. In general, it is an approximation and other XKBA variants have been considered [1, 2, 5]:

$$G^<_x = -G^R_x \rho + \rho G^A_x$$  \hspace{1cm} (46)

Some more important examples are summarized in the table:

<table>
<thead>
<tr>
<th>Abbr.</th>
<th>Ansatz</th>
</tr>
</thead>
<tbody>
<tr>
<td>GKBA</td>
<td>$G^&lt;_G = -G^R_G \rho + \rho G^A_G$  \hspace{1cm} (47)</td>
</tr>
<tr>
<td>QKBA</td>
<td>$G^&lt;_Q = -G^R_Q \rho + \rho G^A_Q$  \hspace{1cm} (48)</td>
</tr>
<tr>
<td>FKBA</td>
<td>$G^&lt;_F = -G^R_F \rho + \rho G^A_F$  \hspace{1cm} (49)</td>
</tr>
</tbody>
</table>

Each Ansatz is specified by a particular choice for the form of the propagators. This is indicated by the label.
Here $G_{R,A}^{R,A}$ denotes the true, fully renormalized propagators, $G_{R}^{R}$ the quasiparticle GF [1, 5] and $G_{0}^{R,A}$ the free particle propagators. When using the Ansatz, all participating GF have to be labeled. Thus, although the GKBAs given by (47) is clearly identical with (45), we use the label ‘G’, so that $G_{G}^{G} \equiv G_{R}^{R}$, etc. It may be asked which use one can expect from approximating the true propagators by another choice. This goes beyond the present discussion, but reasons have been given for using the Quasiparticle Kadanoff-Baym ansatz (QKBA) in particular, in which the propagators are represented by the unrenormalized pole part of the true propagators.

FKBA has been used inadvertently in derivations of the Boltzmann equation or the master equation in the second order weak coupling theory equivalent with the use of the plain Fermi Golden Rule.

Returning to our main task, it is enough to approximate the exact generalized scattering terms in the precursors Quantum Transport Equation (42) by a properly selected Ansatz approximate expression, and the GME for the true particle distribution $\rho$ results:

\[
\frac{\partial \rho}{\partial t} + [H_0, \rho]_\pm = - (\Sigma^R_G \Sigma^<_G - \Sigma^>_G \Sigma^A_G) \\
\quad + (\Sigma^R_G \Sigma^<_A - \Sigma^>_A \Sigma^A_G)
\]

\[
(50)
\]

VII. TRANSPORT EQUATIONS FOR NON-INTERACTING MOLECULAR BRIDGE

Now, we will adopt the structure of the GME derived in the last section for the molecular bridge model and will discuss its suitability for description of dynamics of this model after the first, “wild” period of time evolution, when the initial conditions still matter.

A. GME for the bridge

First of all, let us point out specifications of the molecular bridge model, we will use for to adopt the GME for this model:

First, the system has only one level. This has two consequences: a) the commutator on the l.h.s of the GME vanishes, b) all GF and self-energies are numerical functions of two time arguments, not operators or matrices. The symmetries of the GF components with respect to hermitian conjugation reduce to

\[
G^A(t_1, t_2) = \{G^R(t_2, t_1)\}^* \quad (51)
\]

\[
G^<(t_1, t_2) = - \{G^<(t_2, t_1)\}^* \quad (52)
\]

and analogous relations hold for the self-energies.

Second, the molecular bridge model is a non-selfconsistent model, so that the self-energies including $\Sigma^<$ do not depend on $\rho$ and are not approximated by the Ansatz, as explicitly seen from Eqs. (31), by which the self-energies are given by the lead properties. To sum up,

\[
\Sigma^<_G = \Sigma^A_G
\]

(53)

These properties are enough to obtain the specific GME for the island from Eq. (50) with the use of the Ansatz (46):

\[
\frac{\partial \rho}{\partial t} = - \int _{-\infty}^{t} d\tau \Sigma^R(t, \tau) G^A_A(\tau, t) \cdot \rho(\tau) \\
\quad + \int _{-\infty}^{t} d\tau G^R_A(t, \tau) \Sigma^A_A(\tau, t) \cdot \rho(\tau) \\
\quad - \int _{-\infty}^{t} d\tau \Sigma^<(t, \tau) G^A_A(\tau, t) - \int _{-\infty}^{t} d\tau G^R_A(t, \tau) \Sigma^<(\tau, t)
\]

(54)

\[
(55)
\]

The lower integration limit is symbolically set to $-\infty$, which stands for something like $t_{end}$. The effective lower integration limit saturates much earlier, as it is floating together with $t$, cf. (43). Using the symmetries (52), we bring this to the "final" GME form

\[
\frac{\partial \rho}{\partial t} = - \int _{-\infty}^{t} d\tau 2 \text{Re} \left[ \Sigma^R(t, \tau) G^A_A(\tau, t) \right] \cdot \rho(\tau) \\
\quad - \int _{-\infty}^{t} d\tau 2 \text{Re} \left[ \Sigma^<(t, \tau) G^A_A(\tau, t) \right]
\]

(55)

We use the quotation marks, because this equation has several unusual features and cannot be considered as final. The memory function in the first term is entirely $\rho$-independent as a consequence of the non-interacting model. Thus, the GME is linear in $\rho$. The second, 'backscattering', term does not depend on $\rho$ at all. This is a specific feature of the molecular bridge model caused by its non-selfconsistency, (53). There are no approximations made in this term, as the propagators are not affected by the Ansatz. As a consequence, the propagators in both scattering terms are different: The propagator entering the backscattering is the true one, while the Ansatz related propagator $G^A_A$ appears in the direct scattering term. This second feature has to modified as will be demonstrated now.

Since the molecular bridge model represents a Fermionic quantum system, it is required that the GME could be rewritten in a form expressing the Pauli blocking in scattering integrals explicitly. In the backscattering term, we can write

\[
1 = \rho(\tau) + (1 - \rho(\tau))
\]

(56)
and the whole Eq. (55) can be rearranged to

\[
\frac{\partial \rho}{\partial t} = - \int d\bar{t} 2 \text{Re} [(\Sigma^R(t, \bar{t})G^A_x(\bar{t}, t) + (\Sigma^\lambda(t, \bar{t})G^A(\bar{t}, t)) \cdot \rho(\bar{t})]
\]

As it stands, this equation is not consistent, as the two scattering terms contain different propagators and this leads to a violation of the spectral identity

\[
G^R_x - G^A_x = G^R_x - G^\lambda_x
\]

which, combined with the Ansatz, yields the expected Ansatz expression for \(G^\lambda_x\):

\[
G^\lambda_x = G^R_x(1 - \rho) - (1 - \rho)G^A_x
\]

This can be mended in two ways: either by restricting the Ansatz solely to the original GKBA, or by making the additional Ansatz-like approximation in the back-scattering term in order to give the GME a consistent form,

\[
\frac{\partial \rho}{\partial t} = - \int d\bar{t} 2 \text{Re} [(\Sigma^R(t, \bar{t}) + \Sigma^\lambda(t, \bar{t}))G^A_x(\bar{t}, t)] \cdot \rho(\bar{t})
\]

By the spectral identity, \(\Sigma^R(t, \bar{t}) + \Sigma^\lambda(t, \bar{t}) = \Sigma^\lambda(t, \bar{t}) + \Sigma^A(t, \bar{t})\), but the integration limits take care of the \(\Sigma^A(t, \bar{t})\) term. We accept the more general second alternative: it incorporates the GKBA in a trivial fashion, but it permits to apply another Ansatz as well. Retracing the steps leading to Eq. (60), we arrive at the correspondingly modified bridge GME, as it will be used henceforth as final:

\[
\frac{\partial \rho}{\partial t} = - \int d\bar{t} \left[ \text{Re} \left( \Sigma^R(t, \bar{t})G^A_x(\bar{t}, t) \right) \cdot \rho(\bar{t}) \right] - \int d\bar{t} \frac{\mu_x(t, \bar{t})}{\lambda_x(t, \bar{t})} \left( 1 - \rho(\bar{t}) \right).
\]

The additional modification leading from Eq. (55) to the GME (61) is indicated by an arrow. For the future work with the GME, two correlation functions are defined, as indicated. All physics of the GME is condensed in them.

VIII. FROM GME TO ME

We have seen that the molecular bridge shows a clear tendency to a simpler dynamical structure at times far enough from the last disturbance of the system, when the quantum coherences tend to play lesser role. This leads to the asymptotic non-Markovian dynamical equation (61) of the GME type, which we rewrite here in a convenient form:

\[
\frac{\partial \rho}{\partial t} = - \int d\bar{t} \mu_x(t, \bar{t}) \cdot \rho(\bar{t}) - \int d\bar{t} \lambda_x(t, \bar{t})
\]

where

\[
\mu_x(t, \bar{t}) = 2 \text{Re} \left[ \Sigma^R(t, \bar{t})G^A_x(\bar{t}, t) \right]
\]

\[
\lambda_x(t, \bar{t}) = 2 \text{Re} \left[ \Sigma^\lambda(t, \bar{t})G^A_x(\bar{t}, t) \right]
\]

The natural question arises, if a time interval exists, in which we can proceed further and reduce this GME to a Markovian equation of the general form

\[
\frac{\partial \rho}{\partial t} = I_{\text{IN}}[\rho(t)] - I_{\text{OUT}}(t)
\]

Here, \(I_{\text{IN}}\) is a function, not a functional, so that the equation is time-local, in other words memory-less. The linearity of the GME suggests that the actual form of Eq. (64) will be that of an effective Master Equation (ME). Note that the independence of the backscattering term on the distribution function in (62) is transferred also to Eq. (64).

The GME constructs the present state at each instant \(t\) from the memory of the past. An equivalent transport equation local in time must somehow contract this past into the present. In other words, we meet here the well known problem of moving in the anticausal direction. The feasibility of a derivation and justification of the desired Markovian approximation hinges then again on the assumed property (41) of the self-energies, by which they are confined to a strip of the width \(\tau^*\) in the double-time plane. The same property is then true for the correlation functions (63), and the integrations in (62) thus extend no deeper into the past than \(t - \tau^*\) (cf. Eq. (43)). This suggests approximations of a varying sophistication leading to the reduction of the GME to a time-local form.

A. Simplest approximation - Fermi Golden rule

The limited integration region in the integrals of the GME offers, as the crudest approximation, to neglect the memory effect altogether and to take the density matrix out of the scattering integral, arriving at the following simple linear form of Eq. (64):

\[
\partial_t \rho(t) = \dot{a}_x \rho(t) + \dot{c}_x
\]
with
\[
\tilde{a}_\lambda = \int_{-\infty}^{t} d\tau \mu\lambda(t, \tau), \quad \tilde{c}_\lambda = -\int_{-\infty}^{t} d\tau \lambda\lambda(t, \tau)
\] (66)

Since the Hamiltonian is time-independent presently, we may express the quantities entering the definition (63) of the integrands $\mu\lambda$ and $\lambda\lambda$ by their spectral representation. It follows from Eq. (34) immediately:
\[
G^\xi(t' - t) = +i \int \frac{dE}{2\pi} \tilde{A}_\lambda(E)e^{+iE\tau}\bar{\vartheta}(s) \equiv +i\tilde{A}_\lambda(-s)\vartheta(s)
\]
\[
\Sigma^R(t - t') = -i \int \frac{dE}{2\pi} \Delta^R(E)e^{-iE\tau}\bar{\vartheta}(s) \equiv -i\Gamma(+s)\vartheta(s)
\]
\[
\Sigma^< (t - t') = +i \int \frac{dE}{2\pi} \Delta^<(E)e^{-iE\tau}, \quad s = t - t'
\] (67)

This is obvious for propagators and their self-energies, but in this particular case, also the less self-energy is a single time quantity, because it is controlled by the leads, which are assumed to be stationary. The particle correlation function, by contrast, is time dependent through the time dependent quantity $\rho(t)$ entering the XKB Ansatz.

Using the spectral representation (67) we arrive at the following expressions for the coefficients of the ME (65):
\[
\tilde{a}_\lambda = \int \frac{dE}{2\pi} \Delta(E)\tilde{A}_\lambda(E)
\] (68)
\[
\tilde{c}_\lambda = \int \frac{dE}{2\pi} \Delta^<(E)\tilde{A}_\lambda(E)
\] (69)

To illuminate the meaning of this crude Markovian approximation to the GME, we specialize to the simplest possible Ansatz, the FKBA employing the free propagators. We have:
\[
\partial_t \rho(t) = -\tilde{a}_\lambda \rho(t) + \tilde{c}_\lambda
\] (70)
with the FKBA coefficients (68) and (69) given by
\[
\tilde{A}_\lambda(E) = 2\pi\delta(E - E_b)
\] (71)
\[
\tilde{a}_\lambda = \int \frac{dE}{2\pi} \Delta(E)\tilde{A}_\lambda(E) = \Delta(E_b)
\] (72)
\[
\tilde{c}_\lambda = \int \frac{dE}{2\pi} \Delta^<(E)\tilde{A}_\lambda(E) = \Delta^<(E_b)
\] (73)

Comparing $\Sigma^R$ as given by Eq. (67) and by the explicit partitioning expression quoted in (33), we obtain
\[
\Delta(E) = 2\pi \langle b|H'Q\delta(E - \mathcal{H}_0)QH'|b\rangle
\] (74)
so that Eq. (72) for $\tilde{a}_\lambda$ can be rephrased in words as
\[
\tilde{a}_\lambda = \text{Golden Rule transition rate}
\] (75)

To analyze similarly the $\tilde{c}_\lambda$ coefficient, and to interpret properly the roles of the scattering in and backscattering terms, we have to be more explicit and to write the spectral functions $\Delta^R(E)$ and $\Delta^<(E)$ in a form reflecting the role of both leads. The coupling factors $\alpha_\lambda$ are now constant. Comparing (67) with Eq. (34), we may identify
\[
\Delta^<(E) = \alpha^2_\lambda \Delta^R(E)_{f_R}(E) + \alpha^2_\lambda \Delta^R(E)_{f_R}(E)
\] (76)
Eq. (74) may be similarly rearranged to
\[
\Delta(E) = \alpha^2_\lambda \Delta^R(E) + \alpha^2_\lambda \Delta^R(E)
\] (77)
Using Eqs. (76) and (77) we get
\[
\tilde{c}_\lambda = \alpha^2_\lambda \Delta^R(E_{f_R})_{f_R}(E_b) + \alpha^2_\lambda \Delta^R(E_{f_R})(E_b)
\] (78)
\[
\tilde{a}_\lambda = \alpha^2_\lambda \Delta^R(E_{f_R})(E_b) + \alpha^2_\lambda \Delta^R(E_{f_R})(1 - f_R(E_b))
\] (79)

For $\tilde{a}_\lambda$, this is just a decomposition of (74)–(75) into two separate transition rates to both leads, while for $\tilde{c}_\lambda$, these transition rates for 'backscattering', i.e. feeding of particles from the lead to the island, are shown to depend substantially on the quasi-equilibrium distribution functions of the carriers in each of the leads.

To complete this discussion, we rearrange the ME (65) to a symmetrized form which reflects the Pauli blocking properly and is related to the symmetrized GME (60):
\[
\partial_t \rho(t) = -\tilde{d}_\lambda \rho(t) + b_\lambda(1 - \rho(t))
\] (80)
\[
\tilde{d}_\lambda = \tilde{a}_\lambda - \tilde{c}_\lambda, \quad b_\lambda = \tilde{b}_\lambda = \tilde{b}_0 = \tilde{c}_0
\] (81)

Clearly, $\tilde{d}_\lambda = \Delta^R(E_b)$ is the direct transition rate from the island to the leads incorporating the Pauli blocking, while $\tilde{b}_\lambda = \Delta^<(E_b)$ keeps the meaning of the reverse transition rate back from the leads to the island.

The result of this subsection may be summarized by stating that the true Pauli Master equation is derived from the Generalized Master Equation under two approximate assumptions: first, the memory effect is suppressed completely. The direct transition rate is then given by the integral (72). Second, the propagators are assumed to correspond to free propagation. These two assumptions jointly lead to the transition rate having the form of the Fermi Golden Rule. This result has a parallel in the quantum transport theory, where the Boltzmann limit also requires two combined assumptions: the completed collision approximation (on-shell scattering) and the weak scattering limit.

### B. Linear renormalization

In order to develop a Master Equation going beyond the Fermi Golden Rule, we have to take the memory effect in the GME into account. Using once more the limited integration range of the scattering-in integral, we
extrapolate $\rho(T)$ in the GME (62) back into the past by a linear approximation:
\[ \rho(t) = \rho(t) + \partial_t \rho(t)(\bar{t} - t), \quad t > \bar{t} > t - \tau^* \quad (82) \]
The GME (62) then reduces to the ME
\[ \partial_t \rho(t) = -a_x \rho(t) + c_x \quad (83) \]
where
\[ a_x = \frac{\bar{a}_x}{1 - \bar{a}_x}, \quad c_x = \frac{\bar{c}_x}{1 - \bar{a}_x}, \quad (84) \]
$\bar{a}_x$ and $\bar{c}_x$ are given by (66) and (63), while
\[ \bar{a}_x' = -\int_{-\infty}^{t} d\bar{t} (\bar{t} - t) 2 \mathrm{Re} \left[ \Sigma^R(t, \bar{t}) G_x^A(\bar{t}, t) \right]. \quad (85) \]

In the spectral representation we obtain:
\[ \bar{a}_x = \int \frac{dE}{2\pi} \Delta(E) \bar{A}_x(E) \]
\[ \bar{a}_x' = 2 \int \frac{dE}{2\pi} dE \mathrm{Re} \Sigma(E + i0)(E) \bar{A}_x(E) \quad (86) \]
\[ \bar{c}_x = \int \frac{dE}{2\pi} \Delta^<(E) \bar{A}_x(E) \]

Proceeding similarly as in deriving (80) from (70), we transform Eq. (83) to
\[ \partial_t \rho(t) = -d_x \rho(t) + b_x (1 - \rho(t)) \quad (87) \]
\[ d_x = a_x - c_x, \quad b_x = c_x \]
Structurally, this linearized approximation builds upon the simplest approximation of the GME discussed in the previous section, but modifies it substantially, because the coefficients in the ME are renormalized. The actual role of the renormalization may be seen by inspection of the solution of the ME (83),
\[ \rho(t) = (\rho(t_i) - \rho_{\infty}) e^{-a_x(t-t_i)} + \rho_{\infty}, \quad \rho_{\infty} = \frac{c_x}{a_x} = \frac{\bar{c}_x}{\bar{a}_x} \quad (88) \]

As indicated, the saturation value $\rho_{\infty}$ of the level occupancy does not change by renormalization, as the denominators defined in (84) mutually cancel. The logarithmic decrement $a_x$ of the transition of $\rho$ to steady state is renormalized, however.

The approximation (82) is compatible with any XKB Ansatz. Let us follow the sequence $X = F \rightarrow Q \rightarrow G$. The spectral densities all satisfy the sum rule $\int \frac{dE}{2\pi} \bar{A}_x(E) = 1$. Various $\bar{a}_x$ and $\bar{c}_x$ thus represent weighted averages of the respective spectral densities $\Delta(E)$, $\Delta^<(E)$ with the weight function centered near the island resonant level and evolving as $\delta(E - E_R) \rightarrow$ normalized Lorentzian $\rightarrow$ true $\bar{A}(E)/2\pi$. This different sampling is important, if the spectral densities have pronounced structures.

There is thus a qualitative difference between two limiting spectral profiles, the WBL and the NBL. Clearly $\bar{a}_x$ will be insensitive to the Ansatz in the WBL. The same is true for $\bar{a}_x'$, but on top of that, this quantity will be close to zero except at isolated singular points for WBL. The resulting $a_x$ is then expected to be basically given by the simplest $\bar{a}_x$. By contrast, in the NBL, both $\bar{a}_x$ and $\bar{a}_x'$ are equally important for the decay rate $a_x$ and both are sensitive to the Ansatz dependent spectral sampling in their defining expressions. This contrast WBL vs. NBL is less pronounced for $\bar{c}_x$ and $\bar{c}_x'$, where the spectral inhomogeneity caused by the lead distribution functions makes the integral sensitive to the choice of the Ansatz in both limits.

The gross features of the renormalized decay rate emerge already for FKBA, however, precisely because they are not not obscured by the $\bar{A}$-broadening. The first equation of (84) may be written as
\[ a_x = \frac{2|\mathrm{Im} \Sigma(\bar{E}_b + i0)|}{1 - 2 \mathrm{Re} \frac{\partial}{\partial \Omega} \Sigma(\bar{E}_b + i0)} \quad (89) \]

This is reminiscent of the expression for a quasiparticle energy (presently – renormalized resonant island level). There is a substantial difference, however. The self-energy is doubled at both places. That $|\mathrm{Im} \Sigma|$ is doubled to $2|\mathrm{Im} \Sigma|$ is understandable, because we are dealing with the decay of probabilities, not probability amplitudes. But this explanation does not work for the real part $2\frac{\partial}{\partial \Omega} \mathrm{Re} \Sigma$. It should be noted that this problem appeared long time ago in [48]. There it was tentatively interpreted as another artifact of the KB-like quasi-classical expansion, but here we see it emerging independently of this treatment.

C. Selfconsistent approximation

We will now try to avoid arbitrary assumptions of the two previous approximations by introducing the formal solution of the expected ME into the GME and require its self-consistency.

Let us assume that the GME (55) can be reasonably approximated by a ME with unspecified coefficients,
\[ \partial_t \rho(t) = -a_{\text{rad}} \rho(t) + c_{\text{rad}} \quad (90) \]
starting from some $t_i$. For $t > t_i$, the solution of this ME has the form
\[ \rho(t) = (\rho(t_i) - \rho_\infty) e^{-a_{\text{rad}}(t-t_i)} + \rho_\infty, \quad \rho_\infty = \frac{c_{\text{rad}}}{a_{\text{rad}}} \quad (91) \]
We introduce this explicit solution into the GME (63) and try to adjust the coefficients of the ME (90). By (91), it holds
\[ \rho(T) = (\rho(t_i) - \rho_\infty) e^{-a_{\text{rad}}(T-t_i)} + \rho_\infty \quad (92) \]
for any pair of times $t, \tau > t_i$. Inserting this into the GME (63) yields:

$$\frac{\partial \rho}{\partial t} = - \int_{-\infty}^{t} d\tau \mu_x(t-\tau)e^{-a_{\text{trial}}(\tau-t)} \times (\rho(t) - \rho_{\infty})$$

$$- \int_{-\infty}^{t} d\tau \lambda_x(t-\tau) \times \rho_{\infty} - \int_{-\infty}^{t} d\tau \lambda_x(t-\tau) \times \rho_{\infty} - \int_{-\infty}^{t} d\tau \lambda_x(t-\tau) \times \rho_{\infty}$$

(93)

Using (66) and introducing for $a_{\text{trial}}$ an assumption that this is a root of the equation

$$a_{\text{trial}} = \int_{-\infty}^{t} d\tau \mu_x(t-\tau)e^{-a_{\text{trial}}(\tau-t)} \quad (94)$$

we can rewrite (93) to

$$\frac{\partial \rho}{\partial t} = - a_{\text{trial}} \times (\rho(t) - \rho_{\infty})$$

$$- \tilde{a}_x \times \rho_{\infty} + \tilde{c}_x \quad (95)$$

Comparing (90) with (95), we obtain an identity for $c_{\text{trial}}$

$$c_{\text{trial}} \equiv a_{\text{trial}} \rho_{\infty} - \tilde{a}_x \rho_{\infty} + \tilde{c}_x \quad (96)$$

Combining this identity with $\rho_{\infty} = \frac{c_{\text{trial}}}{a_{\text{trial}}}$ from (91), we get two relations

$$\rho_{\infty} = \frac{\tilde{c}_x}{\tilde{a}_x}, \quad c_{\text{trial}} = \frac{\tilde{c}_x}{\tilde{a}_x} \cdot a_{\text{trial}} \quad (97)$$

The second equation can be rewritten in two ways to demonstrate that both coefficients in the ME are renormalized in the same fashion:

$$a_{\text{trial}} = \frac{\tilde{a}_x}{1 - \frac{\tilde{a}_x}{a_{\text{trial}}}}, \quad c_{\text{trial}} = \frac{\tilde{c}_x}{1 - \frac{\tilde{a}_x}{a_{\text{trial}}}} \quad (98)$$

This is reminiscent of (84), and indeed, expanding the exponential in (94) to terms linear in time, we recover it:

$$\frac{a_{\text{trial}} - \tilde{a}_x}{a_{\text{trial}}} = \frac{1}{a_{\text{trial}}} \int_{-\infty}^{t} d\tau \mu_x(t-\tau) \left[ e^{-a_{\text{trial}}(\tau-t)} - 1 \right]$$

$$\approx \frac{1}{a_{\text{trial}}} \int_{-\infty}^{t} d\tau \mu_x(t-\tau)(-a_{\text{trial}}(\tau-t)) = \tilde{a}_x \tilde{c}_x \quad (99)$$

From here, a direct way leads to the prototypical relation (89) exactly as before; it is rederived, however, without possibly spurious assumptions now.

To clarify the physical meaning of the self-consistent approximation we have to discuss now the integral equation (94). An existence of its root is the basic requirement for validity of the whole approach.

We start our discussion by requiring that the root be positive, $x \leftrightarrow a_{\text{trial}} > 0$, because otherwise the density matrix would diverge for $t \rightarrow \infty$ by (90). Then, however, the exponential in (94) diverges for $t \rightarrow -\infty$. It is then not clear, whether $\mu_x(t-\tau)e^{-z(\tau-t)}$ converges to zero and the integral is well defined. It is true that $\mu_x(t-\tau)$ itself always tends to zero in that limit. In the favorable case, $\mu_x(t-\tau)$ will be negligible beyond the characteristic time $\tau^*$ coinciding in practice with the collision duration time introduced in Eq. (41). In that case, we may estimate that the exponential in (94) grows from unity for $t = \tau$ to a maximum value $\exp(a_{\text{trial}}\tau^*)$ for $t = t - \tau^*$. The basic estimate for the relaxation rate is $a_{\text{trial}} \approx \Delta(E_b)$, cf. (72). The expression in the exponent can thus be rewritten by introducing the relaxation time $1/a_{\text{trial}} \approx 1/\Delta(E_b)$ as $a_{\text{trial}}\tau^* = \tau^*/\tau$. The solubility condition for Eq. (94) assumes the form

$$\frac{\tau^*}{\tau} \leq 1 \quad (100)$$

This inequality is not an additional requirement, but a cornerstone of the whole NGE based theory of quantum transport equations. As discussed at length in [3, 5], the basic prerequisite for using the XKB Ansatz approximation is that a “quasi-particle” picture is valid, in other words the decay time of the current carrying resonant states ($\approx \tau$) is sufficiently long. In particular, it is required that the formation time ($\approx \tau^*$) of the resonant states is shorter (ideally — much shorter) than the decay time. This is, of course, precisely the condition (101). It may be concluded that the GME and an effective ME are governed by the same regime and that the transport controlled by a ME is always achieved as a long time asymptotics of the GME.

This analysis indicates that the self-consistent method C is fully general within the whole Causal Ansatz scheme. It remains to identify the position of the two other approximate methods, B and A. Case B turns out to be specified by the sharper condition $\tau^*/\tau \ll 1$. This is neatly seen from the (99) $\rightarrow$ (100) transition to the linear approximation, for which this strong condition provides a justification and a quantitative criterion. Finally, case A can be obtained from B as a weak scattering limit, that is on a quantitative basis supplementing the hierarchy of characteristic times. The weak scattering enters at two places. First, the propagators in the Ansatz are taken as free. Second, the renormalization denominator in Eq. (89) is set to 1, which means $2|\text{Re} \Sigma_{\text{xx}}(E_b + i0)| \ll 1$, a weak scattering condition. The resulting decay rate then becomes, according to Eq. (74), the Fermi Golden Rule transition probability rate. As pointed out already, only in this double limit, combining the well pronounced quasi-particle behavior with the weak scattering, can the true Pauli Master Equation be recovered.

**IX. CONCLUSIONS**

This concluding section attempts to take a point of view complementary to the main text. There, the model of
a non-interacting molecular bridge was used to explore dynamical behavior of electrons in nanoscopic systems under various regimes with the emphasis on time hierarchies governing transient response of the system and on testing the validity of various approximate concepts in describing the time evolution of the system. Here, we touch upon two general questions. First, we ask about the role of the specific properties of the model, in particular of the complete omission of the interactions. Second, we sum up our view of the finite time initial conditions for a transient process.

**Specific properties of the model**

1. It is not an accident that we can formulate dynamics of the molecular bridge via NGF approach since a non-interacting molecular bridge mimicks pretty well many body systems [6]: a simple excitation island level is coupled to a continuum of double excitations lead states, . . .

2. The simplicity of the molecular bridge model is based on its non-selfconsistent nature. Namely, while the higher order excitations of a true many body system are built up from the simple excitations, here the decay continuum does not involve the island states and is of a completely different nature. This makes the model soluble to the end, because the self-energies are exact already in the second order of the perturbation expansion. It should be noted that this is a generic feature of a whole class of models which can be mapped on the Anderson impurity: the Anderson model itself, Newns-Grimley adsorbate model, Bardeen tunneling Hamiltonian, weak coupling polaron treated in the Tamm-Dankoﬀ model, etc.

3. These two properties of the model are mutually linked and jointly lead to important explicit results. First, the retarded self-energy is entirely specified by the spectral densities $\Delta_l, \Delta_R$, which, in fact play the role of the tunneling functions for both junctions. These two functions are specified beforehand and do not depend on the processes taking place at the island. Their form is decisive for the temporal structure of these processes, however. In particular, the distinction between the wide band limit and the narrow band limit defines the short time behavior of the system and permits to make the Bogolyubov conjecture about the correlations decay quantitatively tested.

In addition, also the $\Sigma<-$ self-energy is given in terms of the $\Delta_{l,R}$ spectral densities and the quasiparticle distribution functions in the leads. Thus, the environment of the island is decisive for the physics of the whole molecular bridge system (as is ostensible already from the Meir-Wingreen formula). This lack of inner dissipative structure of the island is reﬂected in the violation of a local fluctuation-dissipation relationship between $\Sigma^H$ and $\Sigma<$ (which precludes any use of the Kadanoff-Baym Ansatz).

**Treatment of the initial conditions**

4. The issue of complex ($\sim$ correlated) initial conditions at a finite initial time is important both conceptually and practically. They can be accommodated into the general NGF scheme in the two different ways, using either the synchronous, or the diachronous technique. While the diachronous approach is general, the synchronous method depends on the system studied and the approximations used [26, 28]. For the present model, the synchronous solution is exact, and we took the advantage and compared the results, and their meaning, with the diachronous solution. This we develop within the Keldysh real time method: the initial time serves as a joining point between the relevant process and processes preceding it in the past which are extended to a complete Keldysh switch-on process. This has all the advantages of the widespread diachronous method employing an extension to imaginary times [15, 25], while the physical contents is much clearer.

5. The non-interacting molecular bridge model is completely described by its one-particle density matrix. Thus, the full initial condition at any specified time is given in its terms. Because the diagonal blocks are prescribed both in the leads and at the island, the essential information is contained in the off-diagonal island-leads blocks. The transient correction terms in the self-energy of the island state are expressed by means of these off-diagonal blocks – the main qualitative conclusion of the synchronous approach. For the diachronous solution, we employ the time-partitioning method, in which the processes prior to the initial time are linked with the relevant process by the past-future blocks of the self-energy. The efficiency of the method is based on the time structure of the self-energy, whose components are expected to be essentially non-zero only in a restricted strip along the time diagonal (the Bogolyubov principle again). For the present bridge model, the synchronous expressions are fully equivalent with these time-partitioning results [6] and this is the main qualitative conclusion serving for possible generalizations to interacting systems.

6. The construction of the initial states by time partitioning of Keldysh switch-on states has the advantage that all admissible states are "physical", that is can be attained by a real process completed before the relevant process commences. In fact the variety of possible initial states is immense: the initial time may be chosen arbitrarily in the course of a preparatory process. This procedure may be iterated, in particular, if the system undergoes a sequence of abrupt changes separated by comparatively quiescent stages. It is then productive to view the evolution as a series of nested transients.
and to capture the reconstruction of the state of the system by iterating the time partitioning procedure until the final instant of discontinuity which acts to form the stage for the subsequent transition to the kinetic stage and the final equilibration.

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