

## Non-equilibrium stationary states from the equation of motion of open systems

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## Non-equilibrium stationary states from the equation of motion of open systems

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**Abstract.** Non-equilibrium stationary states are solved from the equation of motion of open systems via a Bogoliubov–Born–Green–Kirkwood–Yvon (BBGKY)-like hierarchy. As an example, we demonstrate this approach for the Redfield equation, which is derived from the whole dynamic equation of motion of a central system coupled to two baths through the projector technique. Generalization to other equations of motion is straightforward. For non-interacting central systems, the first equation out of the hierarchy is closed, whereas for interacting systems it is coupled to higher-order equations in the hierarchy. In the case of interacting systems, two systematic approximations, in the form of perturbation theories, are proposed to truncate and solve the hierarchy. A non-equilibrium Wick's theorem is proved to provide a basis for the perturbation theories. As a test of reliability of the proposed methods, we apply them to small systems, where it is also possible to apply other exact direct methods. Consistent results were found.

**Contents**

<b>1. Introduction</b>	<b>2</b>
<b>2. Derivation of a Bogoliubov–Born–Green–Kirkwood–Yvon (BBGKY)-like hierarchy</b>	<b>4</b>
<b>3. Solving the hierarchy</b>	<b>6</b>
3.1. Method 1: starting from equilibrium states . . . . .	7
3.2. Method 2: non-equilibrium cluster expansion . . . . .	10
<b>4. Conclusion and discussion</b>	<b>14</b>
<b>Acknowledgments</b>	<b>14</b>
<b>Appendix A. Perturbation decomposition of <math>\hat{m}</math></b>	<b>14</b>
<b>Appendix B. Proof of non-equilibrium Wick’s theorem</b>	<b>16</b>
<b>Appendix C. Estimation of convergence</b>	<b>17</b>
<b>References</b>	<b>20</b>

**1. Introduction**

A dynamic equation, such as Newton’s equation or Schrödinger’s equation, describes only dynamical processes and, strictly speaking, does not describe evolution towards thermal equilibrium or non-equilibrium stationary states (NESSs). Sometimes, however, it is used to do so together with a presumed thermal equilibrium distribution, such as in the derivation of the Kubo formula of linear response theory [1]. Starting from the dynamic equation and its corresponding Bogoliubov–Born–Green–Kirkwood–Yvon (BBGKY) hierarchy [2], the uniqueness of a stationary solution at thermal equilibrium, in particular the Boltzmann distribution, remains an assumption but not a proved theorem [2]. The Redfield equation [3], on the other hand, puts the description of dynamical evolution, thermal evolution towards the equilibrium state and evolution towards NESSs under a common framework, although it may show an unphysical transient process due to the use of the Markov approximation.

The Redfield equation has long been a standard tool in the study of relaxation processes in the theory of nuclear magnetic resonance [4], optical spectroscopy and chemical dynamical systems [5]. Due to the fact that usually in those studies the central system of interest is modeled by a Hamiltonian with a very low dimension, the lack of an efficient algorithm to solve the Redfield equation, other than direct diagonalization, direct integration and other direct methods based on various propagators [6], is not a serious problem. However, when the Redfield equation is applied to transport calculations, the size of the system is usually much larger. It is then necessary to have a more efficient way to find the NESSs, without which many physical questions remain unanswered.

A famous example is the validity of the phenomenological law of transport. Advancement in science and technology has made it possible to build nanoscale electronic devices [7], where classical phenomenological laws of transport, such as Fourier’s law and Ohm’s law, may not be valid any more [7]. In order to construct a theory of transport for mesoscopic or microscopic systems, and also to check under what circumstances those phenomenological laws hold, one may start from first principles, i.e. the dynamic equation of classical or quantum systems, and add in as few extra assumptions as possible. The Landauer formula [8] makes use of scattering waves from Schrödinger’s equation, but a biased distribution of those waves is assumed to

calculate physical quantities. The non-equilibrium Green's function (NEGF) method starts from two decoupled systems each at their own thermal equilibrium states, which could be very far from the expected NESSs, and treats coupling between the two systems perturbatively [9]. For interacting systems, usually NEGF is used together with the density functional method [10], from which the whole spectrum and corresponding effective wavefunctions are calculated to construct the non-equilibrium density matrix. Both the perturbation and the effective wavefunctions introduce further approximations to the calculation.

The Redfield equation approach to studying transport phenomena is to explicitly couple the system of interest,  $H_S$ , to reservoirs and then use the projector technique [11] to derive an effective equation of motion for the system. One then solves this Redfield equation to get NESSs. The Redfield equation requires validity of the Markovian approximation and treats the coupling between the system and reservoirs within second-order perturbation. This generalization of the Redfield equation to transport studies was first implemented by Saito *et al* [12] and more or less followed by others [13]–[15]. In such cases, one quite often expects to deal with systems with size  $N \approx 100$  in order to be comparable to other methods, such as the Landauer formula and NEGF [7]. This then leads to a Hilbert space with dimension  $2^{100}$ , when we, for example, consider an  $N$ -site chain of spinless fermions with  $N = 100$ , meaning a matrix with dimension  $4^{100}$  in the corresponding Redfield equation. Due to this exponential increase of the problem size and the lack of a more efficient alternative method, currently one can only discuss physical behaviors of very small systems with  $N \approx 10$  [13, 14]. Other propagator-based direct methods [6] make it possible to analyze only slightly larger systems. Conclusions drawn from numerical results for such small systems are not regarded as reliable enough. In this work, we will present a new approach, using the idea of a BBGKY hierarchy to study this kinetic equation. This allows us to develop very efficient and systematic approximate methods to find NESSs. In fact, besides these direct methods, the stochastic wave function method [16] has been generalized and used to solve the Redfield equation [17]. Such stochastic methods are capable of dealing with slightly larger systems, say  $N$  of around 20. It will be an interesting topic of future work to compare the performance of our method against the stochastic methods.

A general Redfield equation can be cast into the following form [13],

$$\frac{\partial \rho(t)}{\partial t} = L_{H_S} \rho(t) + L_{V_{in}} \rho(t) + \lambda^2 L_B(T, \mu) \rho(t) + \lambda^2 L_P(\Delta T, \Delta \mu) \rho(t) \equiv L \rho(t), \quad (1)$$

where  $L_{H_S} \rho = -i[H_S, \rho]$  is the Hamiltonian of the central system. Sometimes we also use  $H_S = H_0 + V$  to separate  $H_S$  into a non-interacting part  $H_0$  and an interaction  $V$ . Finally,  $L_{V_{in}} \rho = -i[V_{in}, \rho]$ , where  $V_{in}$  is a possible induced potential; for example, the electric potential due to charge distribution in the case of charge transport.  $L_B(T, \mu)$  comes from coupling to baths with coupling strength  $\lambda$ , and  $L_P(\Delta T, \delta \mu)$  exists when baths have different temperatures and/or chemical potentials. This equation describes a dynamical process when  $\lambda = 0$ , thermal relaxation towards equilibrium when  $\lambda \neq 0$ ,  $\Delta T = 0$ ,  $\Delta \mu = 0$  and evolution towards NESSs when they are all non-zero.

If we are interested in the long-time steady-state solution  $\rho_\infty$ , then

$$L \rho_\infty = 0, \quad (2)$$

which is sometimes called a stationary Redfield equation. Here,  $L$  is a matrix with dimension  $d^2$ , where  $d$  is the dimension of the system's Hilbert space; for example,  $d = 2^N$  for the  $N$ -site chain of spinless fermions mentioned above. Solving this equation is very costly computationally.

It involves solving a linear system [13] or an eigenvalue system [15] with dimension  $4^N$  plus an eigenvalue system with dimension  $2^N$ . Currently, for interacting central systems, one can usually only solve the Redfield equation numerically up to  $N = 10$  [13]–[15], whereas for non-interacting systems, the equation with  $N$  equal to about 100 can be solved in terms of single-particle Green's functions (GFs) [12]. For example, for non-interacting systems, in [12], a closed equation of single-particle GFs,  $G_1(k^\dagger, k') \equiv \langle c_k^\dagger c_{k'} \rangle = \text{tr}(c_k^\dagger c_{k'} \rho_\infty)$ , was derived from the stationary Redfield equation (2) and solved.

Our idea is basically to extend this GF-based solution of the Redfield equation for non-interacting systems to interacting systems. We consider  $G_1(m^\dagger, m')$ , GFs in the lattice basis, and derive an equation for these GFs. In the presence of interaction, the single-particle GFs  $G_1(m^\dagger, m')$ , generally denoted as  $G_1$ , will be coupled to the two-particle GFs  $G_2(m^\dagger, n^\dagger, m', n') \equiv \langle c_m^\dagger c_n^\dagger c_{m'} c_{n'} \rangle$ , also generally denoted as  $G_2$ , which are then coupled to three-particle GFs,  $G_3(l^\dagger, m^\dagger, n^\dagger, l', m', n') \equiv \langle c_l^\dagger c_m^\dagger c_n^\dagger c_{l'} c_{m'} c_{n'} \rangle$ , also generally denoted as  $G_3$  and so on. We arrive at a BBGKY-like equation hierarchy [2].

In principle, solving the whole hierarchy is as hard as solving the Redfield equation directly. The hierarchy has to be truncated first and then solved. In the rest of this paper, after deriving the hierarchy from the Redfield equation, we will then present two such methods. In the example calculations, we will only truncate the hierarchy at the first equation of the hierarchy. The first method substitutes the value of  $G_2$  at equilibrium for the unknown  $G_2$  appearing in the first equation, whereas the second expresses  $G_2$  as combinations of  $G_1$  via cluster expansion. After either method, the equation is closed and then solved. We will see in the following that both methods are significantly more efficient than the direct methods. The first is capable of dealing with relatively small systems but with large interaction strength, whereas the second can deal with much larger systems but with relatively small interaction strength.

## 2. Derivation of a Bogoliubov–Born–Green–Kirkwood–Yvon (BBGKY)-like hierarchy

For concreteness in presenting our general formulation, let us start with a Redfield equation describing an  $N$ -site chain of spinless fermions coupled with two fermionic baths. Our system of interest is defined by  $H_S$ ,

$$H_S = -t \sum_{l=1}^{N-1} (c_l^\dagger c_{l+1} + c_{l+1}^\dagger c_l) + V_0 \sum_{l=1}^{N-1} c_{l+1}^\dagger c_{l+1} c_l^\dagger c_l = H_0 + V_s. \quad (3)$$

The two heat baths are collections of fermionic modes,

$$\mathcal{H}_B = \sum_{k,\alpha} \omega_{k,\alpha} b_{k,\alpha}^\dagger b_{k,\alpha}, \quad (4)$$

where  $\alpha = L, R$  indexes the left- and right-side baths and we set  $\hbar = 1$ ,  $k_B = 1$ , the lattice constant  $a = 1$  and the hopping constant  $t = 1$ . The system–baths coupling is chosen as

$$V = \lambda \sum_{k,\alpha} V_k^\alpha (c_\alpha^\dagger b_{k,\alpha} + c_\alpha b_{k,\alpha}^\dagger), \quad (5)$$

where the left (right) bath is coupled to the first (last) site:  $c_L = c_1$  and  $c_R = c_N$  and so on. Bath parameters, including temperature and chemical potential, are chosen to be  $(T_L, \mu)$  and  $(T_R, \mu)$  with  $T_{L/R} = T \pm \frac{\Delta T}{2}$ . In the present work, the induced  $L_{V_{in}}$  term in equation (1) is neglected.

The corresponding Redfield equation reads [11, 13]

$$\frac{\partial \rho(t)}{\partial t} = -i[\mathcal{H}_S, \rho(t)] - \lambda^2 \sum_{\alpha=L,R} \{[c_\alpha^\dagger, \hat{m}_\alpha \rho(t)] + [c_\alpha, \hat{m}_\alpha \rho(t)] + \text{h.c.}\}, \quad (6)$$

where  $\hat{m}_L$  ( $\hat{m}_R$ ) is related to  $c_1$  ( $c_N$ ) and  $\hat{m}_L$  ( $\hat{m}_R$ ) is related to  $c_1^\dagger$  ( $c_N^\dagger$ ) [11],

$$\hat{m}_\alpha = \sum_k |V_k^\alpha|^2 \int_0^\infty d\tau c_\alpha(-\tau) e^{-i\omega_{k,\alpha}\tau} \langle 1 - n(\omega_{k,\alpha}) \rangle, \quad (7a)$$

$$\hat{m}_\alpha^\dagger = \sum_k |V_k^\alpha|^2 \int_0^\infty d\tau c_\alpha^\dagger(-\tau) e^{i\omega_{k,\alpha}\tau} \langle n(\omega_{k,\alpha}) \rangle. \quad (7b)$$

Here,  $n(\omega_{k,\alpha}) = (e^{\beta_\alpha(\omega_{k,\alpha} - \mu_\alpha)} + 1)^{-1}$  is the Fermi–Dirac distribution with the bath temperature  $T_\alpha = 1/\beta_\alpha$  and chemical potential  $\mu_\alpha$ . If  $U(t) = e^{-iH_S t}$  is known, then so is  $c_\alpha(t) = U^\dagger(t)c_\alpha U(t)$  and therefore the operators  $\hat{m}$ . This requires a full diagonalization of  $H_S$ . Using eigenvectors of  $H_S$ , one can perform the above integrals to get the operator  $\hat{m}$ s. More details are included in appendix A. There we will also see that a change of variable between summation over  $k$  and integration over energy in equation (7) involves the density of states of the baths  $D_\alpha(\omega)$ . We combine this density of states together with the coupling constant  $V_k^\alpha$ , and set  $D_\alpha(\Omega_{mn})|V_{k_{mn}}^\alpha|^2$  as an overall constant, which is included in  $\lambda^2$  (see appendix A for more details).

When only a long-time steady-state solution  $\rho_\infty$  is of interest, we may derive a stationary form, equation (2), from the kinetic Redfield equation. Furthermore, from equation (2), for a physical quantity of the central system with operator  $A$ , we generally have

$$0 = i\langle [A, H_0] \rangle + i\langle [A, V] \rangle + \lambda^2 \sum_\alpha \{ \langle [A, c_\alpha^\dagger] \hat{m}_\alpha \rangle + \langle [A, c_\alpha] \hat{m}_\alpha^\dagger \rangle - \langle \hat{m}_\alpha^\dagger [A, c_\alpha] \rangle - \langle \hat{m}_\alpha [A, c_\alpha^\dagger] \rangle \}, \quad (8)$$

where  $\hat{m}_\alpha^\dagger$  ( $\hat{m}_\alpha^\dagger$ ) is the Hermitian conjugate of  $\hat{m}_\alpha$  ( $\hat{m}_\alpha$ ). All equations of GFs in the rest of this paper will be derived from this equation. For example, the first and second equation of the hierarchy can be derived by using  $A = c_m^\dagger c_n$  and  $A = c_m^\dagger c_n^\dagger c_{m'} c_{n'}$  in equation (8),

$$0 = it \langle c_{m-1}^\dagger c_n \rangle + it \langle c_{m+1}^\dagger c_n \rangle - it \langle c_m^\dagger c_{n+1} \rangle - it \langle c_m^\dagger c_{n-1} \rangle \quad (9a)$$

$$-iV_0 \langle c_m^\dagger c_{n-1}^\dagger c_n c_{n-1} \rangle + iV_0 \langle c_{m+1}^\dagger c_m^\dagger c_{m+1} c_n \rangle - iV_0 \langle c_{n+1}^\dagger c_m^\dagger c_{n+1} c_n \rangle + iV_0 \langle c_m^\dagger c_{m-1}^\dagger c_n c_{m-1} \rangle \quad (9b)$$

$$- \lambda^2 \sum_\alpha \langle \delta_{m\alpha} c_n \hat{m}_\alpha + \delta_{n\alpha} \hat{m}_\alpha^\dagger c_m^\dagger - \delta_{n\alpha} c_m^\dagger \hat{m}_\alpha - \delta_{m\alpha} \hat{m}_\alpha^\dagger c_n \rangle \quad (9c)$$

and

$$0 = it \langle c_m^\dagger c_n^\dagger c_{m'} c_{n'+1} \rangle + it \langle c_m^\dagger c_n^\dagger c_{m'} c_{n'-1} \rangle + it \langle c_m^\dagger c_n^\dagger c_{m'+1} c_{n'} \rangle + it \langle c_m^\dagger c_n^\dagger c_{m'-1} c_{n'} \rangle \\ - it \langle c_m^\dagger c_{n-1}^\dagger c_{m'} c_{n'} \rangle - it \langle c_m^\dagger c_{n+1}^\dagger c_{m'} c_{n'} \rangle - it \langle c_{m-1}^\dagger c_n^\dagger c_{m'} c_{n'} \rangle - it \langle c_{m+1}^\dagger c_n^\dagger c_{m'} c_{n'} \rangle \\ + iV_0 \langle c_m^\dagger c_n^\dagger c_{m'} c_{n'} \rangle (\delta_{m'+1,n'} + \delta_{m'-1,n'} - \delta_{m+1,n} - \delta_{m-1,n}) \quad (10a)$$

$$- iV_0 \sum_{l=m\pm 1, n\pm 1} \langle c_l^\dagger c_m^\dagger c_n^\dagger c_l c_{m'} c_{n'} \rangle + iV_0 \sum_{l=m'\pm 1, n'\pm 1} \langle c_l^\dagger c_m^\dagger c_n^\dagger c_l c_{m'} c_{n'} \rangle \quad (10b)$$

$$\begin{aligned}
& -\lambda^2 \sum_{\alpha} \left\langle \delta_{m'\alpha} c_m^{\dagger} c_n^{\dagger} c_{n'} \hat{m}_{\alpha} - \delta_{n'\alpha} c_m^{\dagger} c_n^{\dagger} c_{m'} \hat{m}_{\alpha} + \delta_{m\alpha} c_n^{\dagger} c_{m'} c_{n'} \hat{m}_{\alpha} - \delta_{n\alpha} c_m^{\dagger} c_{m'} c_{n'} \hat{m}_{\alpha} \right\rangle \\
& -\lambda^2 \sum_{\alpha} \left\langle \delta_{n'\alpha} \hat{m}_{\alpha}^{\dagger} c_m^{\dagger} c_n^{\dagger} c_{m'} - \delta_{m'\alpha} \hat{m}_{\alpha}^{\dagger} c_m^{\dagger} c_n^{\dagger} c_{n'} + \delta_{n\alpha} \hat{m}_{\alpha}^{\dagger} c_m^{\dagger} c_{m'} c_{n'} - \delta_{m\alpha} \hat{m}_{\alpha}^{\dagger} c_n^{\dagger} c_{m'} c_{n'} \right\rangle.
\end{aligned} \tag{10c}$$

Note that since the set of all polynomials of  $\{c_l, c_l^{\dagger}\}$  form a complete basis of the operator space, operators  $\hat{m}$  are certain functions of polynomials of  $\{c_l, c_l^{\dagger}\}$ . Therefore, as expected,  $G_1$  is coupled to  $G_2$  from equation (9b), and possibly also  $G_3$  or higher GFs from equation (9c), and  $G_2$  is coupled to  $G_3$  from equation (10b), and possibly also  $G_4$  or higher GFs from equation (10c). Solving such an equation hierarchy is not easier than directly solving the Redfield equation, unless  $V_0 = 0$ , so that the above equation of  $G_1$  is closed and is not coupled to  $G_2$ .

We may, however, solve these equations by truncating the hierarchy at a certain order with some further approximations, such as the molecular-chaos assumption in the classical Boltzmann equation [11], or replacing high-order GFs by cluster expansion of lower-order ones [18, 19]. In this work, we suggest the following two approximate methods: (i) substitution of certain high-order GFs by their values at equilibrium; and (ii) expression of high-order GFs as combinations of lower-order ones plus a correlation part via cluster expansion and then ignoring the correlation part at a certain order. Specifically in the following example calculation, the first-order form of both approximations (i.e. only the first equation of the hierarchy) is used and substitution or cluster expansion is performed on  $G_2$ . One can do such a substitution or cluster expansion of GFs at further-order GFs and make use of further equations in the hierarchy. A general discussion of the accuracy of such substitutions at different orders will be presented elsewhere. In this work, we focus on the potential of this BBGKY-like formulation and discuss briefly the topic of performance of the two approximations in their first-order forms.

### 3. Solving the hierarchy

In order to solve equation (9) explicitly, we will first have to find explicit forms of operators  $\hat{m}$  in terms of operator  $\{c_l, c_l^{\dagger}\}$ . In the following, we will present one exact numerical calculation and one perturbative calculation of those operators. Correspondingly, based on these two methods of finding operators  $\hat{m}$ , we will discuss in this section two ways of making equation (9) a closed equation by dealing with  $G_2$  terms in the equation differently.

We will first discuss a more accurate method, even for a large  $V_0$ , but computationally costly: perturbation based on two-particle GFs at equilibrium. Next we will discuss a relatively less accurate but computationally much cheaper method, the non-equilibrium cluster expansion. The latter works only for relatively small  $V_0$ , but it can be applied to much larger systems. The unknown  $G_1(m^{\dagger}, n)$  solved from both methods will be compared against  $G_1^{\text{Ex}}(m^{\dagger}, n)$ , the exact solution of equation (2). A measure of the relative distance between two matrices  $A$  and  $B$ ,

$$d_B^A = \frac{\sqrt{\sum_{ij} |A_{ij} - B_{ij}|^2}}{\sqrt{\sum_{ij} |B_{ij}|^2}}, \tag{11}$$

is used to describe the accuracy of our approximations.

### 3.1. Method 1: starting from equilibrium states

As explicitly worked out in equation (A.2) in appendix A, operators  $\hat{m}$  can be written in eigenmodes of  $H_S$ , which can be solved from an exact diagonalization of  $H_S$ , a  $2^N$ -dimension eigenvalue problem. Then in the language of super-operator space [15], where operators are treated like vectors (so-called super-vectors), super-vectors  $\hat{m}$  can be expanded on the basis of polynomials of  $\{c_l, c_l^\dagger\}$ ,

$$\hat{m}_\alpha = \sum_l d_{\alpha;l} c_l + V_0 D_\alpha, \quad (12a)$$

$$\hat{\bar{m}}_\alpha = \sum_l \bar{d}_{\alpha;l} c_l^\dagger + V_0 \bar{D}_\alpha. \quad (12b)$$

Here we keep only the linear polynomial in the present work, although further expansion is possible. Using the definition of the inner product between super-vectors  $\langle\langle A|B \rangle\rangle = \text{tr}(A^\dagger B)$ , we have

$$d_{\alpha;l} = \frac{1}{2^{(N-1)}} \text{tr}(c_l^\dagger \hat{m}_\alpha), \quad (13a)$$

$$\bar{d}_{\alpha;l} = \frac{1}{2^{(N-1)}} \text{tr}(c_l \hat{\bar{m}}_\alpha), \quad (13b)$$

and operators  $V_0 D$  and  $V_0 \bar{D}$  are just the remaining part of operators  $\hat{m}$  and  $\hat{\bar{m}}$ , respectively. Here,  $2^{N-1}$  is a normalization constant to make  $d_{\alpha,l} = 1$  when  $\hat{m}_\alpha = c_l$ .

With the above expressions for operators  $\hat{m}$ , equation (9) becomes

$$0 = \text{it}\langle c_{m-1}^\dagger c_n \rangle + \text{it}\langle c_{m+1}^\dagger c_n \rangle - \text{it}\langle c_m^\dagger c_{n+1} \rangle - \text{it}\langle c_m^\dagger c_{n-1} \rangle \\ + \lambda^2 \sum_{l,\alpha} \langle \delta_{n\alpha} (d_{\alpha;l} + \bar{d}_{\alpha;l}^*) c_m^\dagger c_l + \delta_{m\alpha} (\bar{d}_{\alpha;l} + d_{\alpha;l}^*) c_l^\dagger c_n \rangle \quad (14a)$$

$$- \lambda^2 \sum_\alpha [\delta_{m\alpha} \bar{d}_{\alpha;n} + \delta_{n\alpha} \bar{d}_{\alpha;m}^*] \quad (14b)$$

$$- iV_0 \langle c_m^\dagger c_{n-1}^\dagger c_n c_{n-1} \rangle + iV_0 \langle c_{m+1}^\dagger c_m^\dagger c_{m+1} c_n \rangle - iV_0 \langle c_{n+1}^\dagger c_m^\dagger c_{n+1} c_n \rangle + iV_0 \langle c_m^\dagger c_{m-1}^\dagger c_n c_{m-1} \rangle \quad (14c)$$

$$- \lambda^2 V_0 \sum_\alpha \langle \delta_{m\alpha} c_n \bar{D}_\alpha + \delta_{n\alpha} \bar{D}_\alpha^\dagger c_m^\dagger - \delta_{n\alpha} c_m^\dagger D_\alpha - \delta_{m\alpha} D_\alpha^\dagger c_n \rangle. \quad (14d)$$

Note that every  $c_0, c_0^\dagger, c_{N+1}$  and  $c_{N+1}^\dagger$  that appears in the equation should be recognized as 0. First, let us replace all  $G_2$ s in equation (14c) by their values at equilibrium, denoted here as  $G_2^{E,(0)}$  where the superscripts indicate the usage of thermal equilibrium (denoted by superscript  $E$ ) as the zeroth-order approximation (denoted by (0)) of the non-equilibrium  $G_2$ . Using the first term as an example,

$$G_2^{E,(0)}(m^\dagger, n) = \text{tr}(c_m^\dagger c_{n-1}^\dagger c_n c_{n-1} \rho_{\text{eq}}(H_S)), \quad (15)$$

where  $\rho_{\text{eq}}(H_S) = \frac{1}{Z} e^{-H_S/T}$ . This requires eigenstates of  $H_S$ . Similarly, one can define  $G_D^{E,(0)}$  from equation (14d), using the first term as an example,

$$G_D^{E,(0)}(m^\dagger, n) = \delta_{m\alpha} \text{tr}(c_n \bar{D}_\alpha \rho_{\text{eq}}(H_S)). \quad (16)$$

Next, let us calculate  $G_1^{E,(1)}$  from equation (14), where the superscript (1) means that the approximate calculation takes care of the first equation of the hierarchy, equation (14). We organize all  $G_1^{E,(1)}(m^\dagger, n)$  as a vector,

$$g_1^{E,(1)} = [G_1(1^\dagger, 1), G_1(1^\dagger, 2), \dots, G_1(N^\dagger, N)]^T, \quad (17)$$

then equation (14) for given values of  $m, n$  is the equation occupying the  $(mN+n)$ th row and totally there are  $N^2$  such equations. After substituting  $G_2^{E,(0)}$  and  $G_D^{E,(0)}$  for the exact but unknown  $G_2$  and  $G_D$ , the whole set of equation (14) for all  $m, n$  then becomes a linear system on  $g_1^{E,(1)}$  with dimension  $N^2$ ,

$$\Gamma^{(1)} g_1^{E,(1)} = iV_0 g_2^{E,(0)} + \lambda^2 \nu + \lambda^2 V_0 g_D^{E,(0)}, \quad (18)$$

where the vector  $\nu$  comes from ordering equation (14b) in the same way as  $g_1^{E,(1)}$ . The same holds for  $g_2^{E,(0)}$  and  $g_D^{E,(0)}$  correspondingly from ordering equations (14c) and (14d), and from equation (14a) one gets the matrix  $\Gamma^{(1)}$ . For example, assuming  $m$  and  $n$  are not at boundaries, one may write from equation (14)

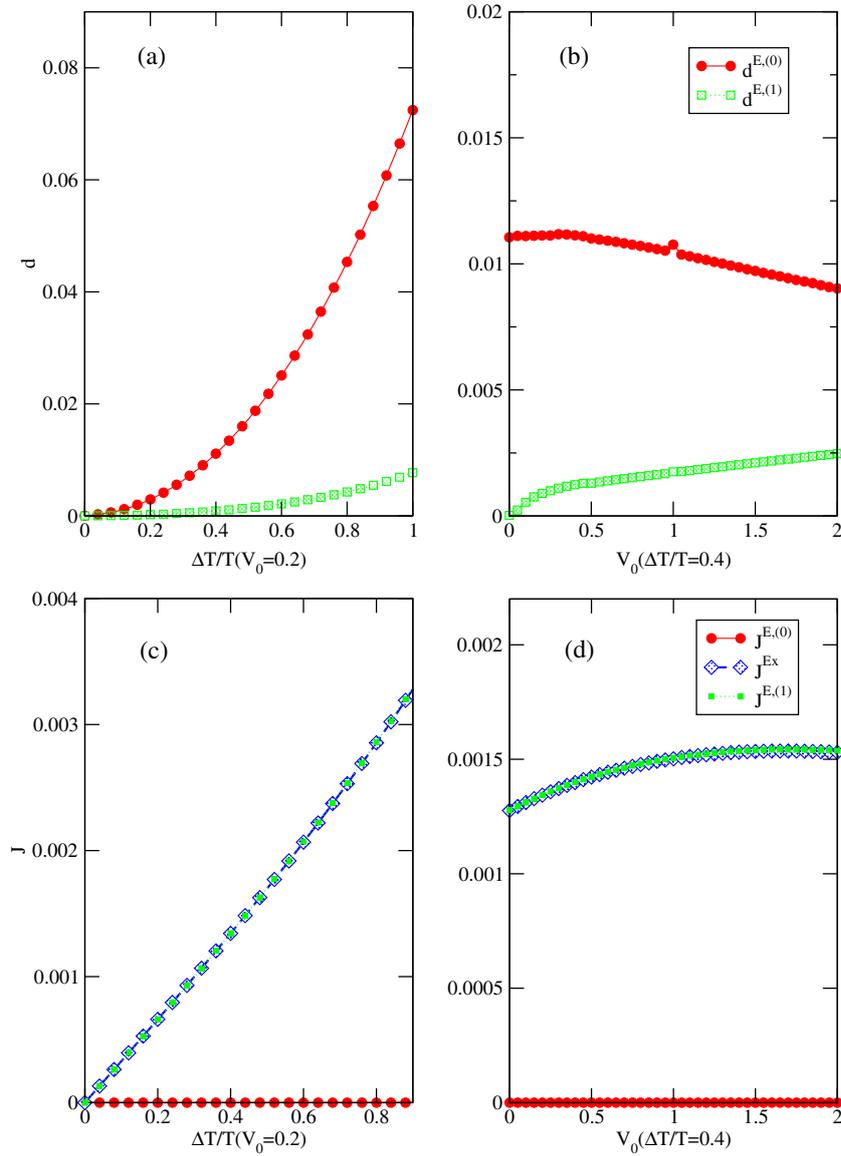
$$\nu_{mN+n} = \sum_{\alpha} [\delta_{m\alpha} \bar{d}_{\alpha;n} + \delta_{n\alpha} \bar{d}_{\alpha;m}^*], \quad (19a)$$

$$\Gamma_{(mN+n),((m-1)N+n)}^{(1)} = i t. \quad (19b)$$

Next, we calculate single-particle equilibrium GFs,  $G_1^{E,(0)}$ , and organize it in the same way into a vector denoted as  $g_1^{E,(0)}$ . In order to set a reference of the accuracy, we compare  $d^{E,(0)}$ , the difference between the exact solution  $g_1^{\text{Ex}}$  and the zeroth order  $g_1^{E,(0)}$ , and  $d^{E,(1)}$ , the difference between the exact solution  $g_1^{\text{Ex}}$  and the first order solution above,  $g_1^{E,(1)}$ . Here,  $G^{\text{Ex}}(m^\dagger, n) = \text{tr}(c_m^\dagger c_n \rho_\infty)$ , where  $\rho_\infty$  is the exact solution from equation (2).

**3.1.1. Results.** Firstly, we set  $V_0 = 0.2$  as a constant, and check the accuracy of  $g_1^{E,(1)}$  with different values of  $\Delta T$ . From figure 1(a), we can see that the worst case is about  $d^{(1)} = 1\%$ . Secondly, we set  $\Delta T = 0.4T$  as a constant, and check the accuracy of  $g_1^{E,(1)}$  with different values of  $V_0$ . The worst case is  $d^{(1)} = 0.3\%$ , as shown in figure 1(b). Overall,  $d^{E,(1)}$  is always much smaller than  $d^{E,(0)}$ .  $J^{E,(0)}$ ,  $J^{E,(1)}$  and  $J^{\text{Ex}}$  are calculated, respectively, from  $g_1^{E,(0)}$ ,  $g_1^{E,(1)}$  and  $g_1^{\text{Ex}}$ . From figure 1(c) and (d) we see that in both cases,  $J^{E,(1)}$  is very close to the exact,  $J^{\text{Ex}}$ , while  $J^{E,(0)}$ , the current in the equilibrium state, is always zero. Very high accuracy is found, especially for small  $\Delta T$ . This indicates that the approximation captures the essential part of the NESSs. It is also worth mentioning that this method generates reasonable results for very large  $V_0$ . Furthermore, it is likely that the approximation could be improved: from further expansions in terms of higher-order polynomials of  $c_l, c_l^\dagger$  and substitution of their values at equilibrium for the higher-order unknown GFs in higher-order equations of the hierarchy. Stopping the expansion of operators  $\hat{m}$  at linear order of  $V_0$  is compatible with solving only the first equation of the hierarchy. If further equations of the hierarchy are used, then one should also expand operators  $\hat{m}$  in further orders of  $V_0$ .

In order to estimate the accuracy of the first-order form of this approximation, and also to get an overview of the accuracy of possibly the next order, let us study the leading order of residues in terms of  $\lambda^2$  and  $\frac{\Delta T}{T}$ , which are assumed to be small in the following. Hence  $\lambda^2 V_0 \ll V_0$ ; therefore we know that  $g_D$  is relatively smaller than the other  $g_2$  term, so we drop



**Figure 1.**  $g_1^{E,(1)}$  is compared with  $g_1^{Ex}$  for interacting systems at non-equilibrium. (a)  $V = 0.2$ ,  $d^{E,(1)}$  is compared with the reference  $d^{E,(0)}$  for different values of  $\Delta T$ . With larger  $\Delta T$ ,  $d^{(1)}$  becomes larger but is still much smaller than  $d^{E,(0)}$ . (b)  $\Delta T = 0.4T$ ; accuracy was checked for different values of  $V_0$ . In the worst cases shown in the plot,  $d^{(1)}$  is about 0.3%, where  $V_0 = 2t$  is a relatively large strength of interaction. Electrical currents  $J^{E,(0)}$ ,  $J^{E,(1)}$  are compared against  $J^{Ex}$  in (c) and (d). We see that  $J^{E,(0)}$  is zero while  $J^{E,(1)}$  is close to  $J^{Ex}$  for even relatively large  $V_0$ . In all these example calculations,  $t = 1.0$ ,  $\lambda = 0.1$  and  $\mu = -1.0$ .

it. This in fact requires  $\lambda^2 V_0 \ll T$ , which we assume to be true. Similarly, for the same reason, since  $\lambda^2 \Delta T \ll \Delta T$ , we drop the  $\lambda^2 \Delta T$  term in  $\lambda^2 \nu$  in equation (18),

$$\lambda^2 \nu = \lambda^2 \nu_0(T) + \lambda^2 \Delta T \nu_{,T}, \quad (20)$$

and keep only the major term,  $\lambda^2 v_0(T)$ , which is independent of  $\Delta T$ . Here,  $v_{,T}$  denotes formally a derivative of  $T$  on  $v$ . The general idea is then to write down, respectively, equations for  $g_1^{\text{Ex}}$  and  $g_1^{E,(1)}$ , and then compare the two equations to estimate  $\Delta_1^{E,(1)} = g_1^{E,(1)} - g_1^{\text{Ex}}$ . In order to get some information about how such an approximation at the next order improves the accuracy, we also want to compare  $\Delta_1^{E,(1)}$  to  $\Delta_1^{E,(0)} = g_1^{E,(0)} - g_1^{\text{Ex}}$ , which is estimated in the same way from the difference between the equations, respectively, for  $g_1^{\text{Ex}}$  and  $g_1^{E,(0)}$ . See appendix C for further details of those equations and the estimation. Here we summarize the results that

$$\Delta_1^{E,(0)} = \Delta T (\Gamma_0^{(1)})^{-1} \Gamma_{,T}^{(1)} g_1^{\text{Ex}} + iV_0 (\Gamma_0^{(1)})^{-1} \Delta_2^{E,(0)} \quad (21)$$

and

$$\Delta_1^{E,(1)} = (\Gamma_0^{(1)})^{-1} [-V_0^2 (\Gamma_0^{(2)})^{-1} \Delta_3^{E,(0)} + iV_0 \Delta T (\Gamma_0^{(2)})^{-1} \Gamma_{,T}^{(2)} g_2^{\text{Ex}} + iV_0 \lambda^2 (\Gamma_0^{(2)})^{-1} \Delta_1^{E,(0)}]. \quad (22)$$

Here,  $\Delta_n^{E,(0)} = g_n^{E,(0)} - g_n^{\text{Ex}}$  and  $\Delta_n^{E,(1)} = g_n^{E,(1)} - g_n^{\text{Ex}}$ . We refer the readers to appendix C for definitions of all  $\Gamma$  matrices. Most importantly, here we see that  $\Delta_1^{E,(0)}$  is multiplied by a small number  $\lambda^2 V_0$  and then becomes a part of  $\Delta_1^{E,(1)}$ . Furthermore, this relation holds generally for higher-order forms of this approximation. Judging from this term, as long as  $\lambda^2 V_0$  is a small number compared with  $t$ , the method under consideration is very reasonable. As for the other two additional terms, they can be regarded as  $(V_0^2 g_3^{\text{Ex}} + V_0 g_2^{\text{Ex}}) \Delta T$ . Therefore, the limit of  $\frac{V_0}{t}$  where this method is still applicable goes as  $|g_2^{\text{Ex}}|^{-1}$  or  $|g_3^{\text{Ex}}|^{-1/2}$ , which could be much larger than 1 since roughly  $|g_n^{\text{Ex}}| = |g^{\text{Ex}}|^n$ —smaller for larger  $n$ . This explains why, as we see from figure 1, this method is applicable even for  $V_0$  larger than  $t$ . We have also tested several systems with larger  $N$  (up to  $N = 8$ ) and no qualitative difference in accuracy has been found. More details and a more systematic analysis will be presented elsewhere.

### 3.2. Method 2: non-equilibrium cluster expansion

Another way to make equation (9) a closed equation is to use cluster expansion. In the case of equilibrium GFs, it proposes, for example, at the level of two-particle GFs,

$$G_2(m^\dagger, n^\dagger, m', n') = -G_1(m^\dagger, m') G_1(n^\dagger, n') + G_1(m^\dagger, n') G_1(n^\dagger, m') + \mathfrak{G}_2(m^\dagger, n^\dagger, m', n'), \quad (23a)$$

and then sets

$$\mathfrak{G}_2 = 0. \quad (24)$$

It can be applied to higher-order GFs, for example, by a similar expansion on  $G_3$  and setting  $\mathfrak{G}_3 = 0$ . The fact that the equilibrium Wick's theorem shows that indeed  $\mathfrak{G}_2 = 0$  when  $V_0 = 0$  makes this expansion plausible for equilibrium GFs. Here, in the case of NEGFs, we are going to propose the same expansion, and this requires a non-equilibrium Wick's theorem to be true so that  $\mathfrak{G}_2 = 0$  when  $V_0 = 0$  holds for NEGFs. Fortunately, this can be proved (see appendix B for complete details). Setting  $\mathfrak{G}_2 = 0$  with  $V_0 \neq 0$  is similar to using the Hartree–Fock approximation, so that, depending on the system and the physical problem under investigation, one may quite often need to go beyond that to the next level of approximation, i.e. keeping  $\mathfrak{G}_2$  but ignoring  $\mathfrak{G}_3$ , and truncating the equation hierarchy at the second equation instead of the first equation. In this work, we will use the first-level approximation, i.e. ignoring  $\mathfrak{G}_2$ .

However, with operators  $D$  defined in the previous section, cluster expansion cannot be applied; instead we have to expand operators  $\hat{m}$  in higher-order polynomials of  $\{c_l, c_l^\dagger\}$ . This can be done as follows. Other than the exact direct diagonalization, operators  $\hat{m}$  can also be found perturbatively analytically (see appendix A for full details). The basic idea is to start by assuming

$$c_l(t) = c_l^{(0)}(t) + V_0 c_l^{(1)}(t) + O(V_0^2), \quad (25)$$

and then to derive and solve the equations of motion of  $c_l^{(0)}, c_l^{(1)}$  from Heisenberg's equation. In this way, one avoids the direct diagonalization of  $H_S$  so that it simplifies the calculation, but its accuracy depends on the order of  $V_0$  at which the expansion stops. Stopping at the linear order of  $V_0$  is compatible with the cluster expansion at  $G_2$ . If cluster expansion at higher-order GFs is applied, then operators  $\hat{m}$  should also be expanded in higher orders of  $V_0$ . Keeping only the first order, operators  $\hat{m}$  become

$$\hat{m}_\alpha = \sum_m \mathfrak{D}_{\alpha;m} c_m + V_0 \sum_{m_1 m_2 m_3} \mathfrak{D}_{\alpha;m_1 m_2 m_3} c_{m_1} c_{m_2}^\dagger c_{m_3} + O(V_0^2), \quad (26a)$$

$$\hat{m}_\alpha^\dagger = \sum_m \bar{\mathfrak{D}}_{\alpha;m} c_m^\dagger - V_0 \sum_{m_1 m_2 m_3} \mathfrak{D}_{\alpha;m_1 m_2 m_3} c_{m_3}^\dagger c_{m_2} c_{m_1} + O(V_0^2), \quad (26b)$$

where definitions of  $\mathfrak{D}_{\alpha;m}$  and  $\mathfrak{D}_{\alpha;m_1 m_2 m_3}$  are given in appendix A.

With the first-order cluster expansion and the above expansion of operators  $\hat{m}$  plugged into equation (9), we have

$$0 = i t G_1(m-1; n) + i t G_1(m+1; n) - i t G_1(m; n+1) - i t G_1(m; n-1) \\ + \lambda^2 \sum_{l, \alpha} [\delta_{n\alpha} (\mathfrak{D}_{\alpha;l} + \bar{\mathfrak{D}}_{\alpha;l}^*) G_1(m; l) + \delta_{m\alpha} (\bar{\mathfrak{D}}_{\alpha;l} + \mathfrak{D}_{\alpha;l}^*) G_1(l; n)] \quad (27a)$$

$$+ \lambda^2 V_0 \sum_{\alpha, m_1, m_2} (\mathfrak{D}_{\alpha; n m_2 m_1} - \mathfrak{D}_{\alpha; m_1 m_2 n}) G_1(m_1, m_2) \delta_{m\alpha} \\ + \lambda^2 V_0 \sum_{\alpha, m_1, m_2} (\mathfrak{D}_{\alpha; m m_2 m_1} - \mathfrak{D}_{\alpha; m_1 m_2 m}) G_1(m_2, m_1) \delta_{n\alpha} \quad (27b)$$

$$- \lambda^2 \sum_{\alpha} (\delta_{m\alpha} \bar{\mathfrak{D}}_{\alpha;n} + \delta_{n\alpha} \bar{\mathfrak{D}}_{\alpha;m}^*) \quad (27c)$$

$$+ \lambda^2 V_0 \sum_{\alpha, m_1} (\mathfrak{D}_{\alpha; m_1 m_1 n} \delta_{m\alpha} + \mathfrak{D}_{\alpha; m_1 m_1 m} \delta_{n\alpha}) \quad (27d)$$

$$- i V_0 G_1(m; n-1) G_1(n-1; n) + i V_0 G_1(m; n) G_1(n-1; n-1) \\ - i V_0 G_1(m+1; m+1) G_1(m; n) + i V_0 G_1(m+1; n) G_1(m; m+1) \\ - i V_0 G_1(n+1; n) G_1(m; n+1) + i V_0 G_1(n+1; n+1) G_1(m; n) \\ - i V_0 G_1(m; n) G_1(m-1; m-1) + i V_0 G_1(m; m-1) G_1(m-1; n). \quad (27e)$$

Next we define a vector  $g_1^{C,(1)}$ , where superscript  $C$  means cluster expansion and (1) symbolizes keeping only the first equation in the hierarchy, similarly to  $g_1^{E,(1)}$ . For simplicity of expressions,

let us order equation (27e) in the same way and denote it as  $g_2^{C,(1)} = \Pi(g_1^{C,(1)})$ , where  $\Pi$  refers to the nonlinear function—summation of product—of  $g_1^{C,(1)}$  in equation (27e). Then the above equation can be denoted as

$$(\Gamma_0^{(1)} + \lambda^2 V_0 \Gamma_D^{(1)}) g_1^{C,(1)} = \lambda^2 \nu_0 + \lambda^2 V_0 \nu_1 + i V_0 g_2^{C,(1)}, \quad (28)$$

where the five terms are, respectively, the five subequations in equation (27); for example,

$$(\nu_0)_{mN+n} = \sum_{\alpha} (\delta_{m\alpha} \bar{\mathfrak{D}}_{\alpha;n} + \delta_{n\alpha} \bar{\mathfrak{D}}_{\alpha;m}^*). \quad (29)$$

This equation can be solved iteratively,

$$g_1^{(n+1)} = (\Gamma_0^{(1)} + \lambda^2 V_0 \Gamma_D^{(1)})^{-1} (\lambda^2 \nu_0 + \lambda^2 V_0 \nu_1 + i V_0 \Pi(g_1^{(n)})), \quad g_1^{(0)} = (\Gamma_0^{(1)})^{-1} \lambda^2 \nu_0, \quad (30)$$

where we start from  $g_1^{(0)} = g_1^{C,(0)}$ , which is the exact solution of equation (28) when  $V_0 = 0$ . Through the iteration defined above, we get the solution  $g_1^{C,(1)} = \lim_{n \rightarrow \infty} g_1^{(n)}$ , which in practice stops at large enough  $n$  such that  $g_1^{(n)} - g_1^{(n-1)}$  is small enough.

**3.2.1. Results.** In a similar manner, we define  $\Delta_1^{C,(0)}$  ( $d^{C,(0)}$ ) as the absolute (relative) distance between  $g_1^{C,(0)}$  and  $g_1^{\text{Ex}}$ , and  $\Delta_1^{C,(1)}$  ( $d^{C,(1)}$ ) as the absolute (relative) distance between  $g_1^{C,(1)}$  and  $g_1^{\text{Ex}}$ . Firstly, we set  $V_0 = 0.2$  as a constant, and check the accuracy of  $g_1^{C,(1)}$  with different values of  $\Delta T$ . From figure 2(a) we can see that the worst case is around  $d^{(1)} = 1\%$ . Secondly, we set  $\Delta T = 0.4T$  as a constant, and check the accuracy of  $g_1^{C,(1)}$  with different values of  $V_0$ . The worst case is around  $d^{(1)} = 2\%$ , as shown in figure 2(b). Overall,  $d^{C,(1)}$  is always much smaller than  $d^{C,(0)}$ . From figure 2(c), we see that for a small  $V_0$ ,  $J^{C,(0)}$  already provides a major contribution. However, in figure 2(d) when  $V_0$  becomes larger, the difference between  $J^{C,(1)}$  and  $J^{C,(0)}$  becomes more important. We should also note that for larger  $V_0$ ,  $J^{C,(1)}$  starts to deviate from  $J^{\text{Ex}}$ . This indicates that the approximation captures the essential part of the interaction, but it is more accurate for small  $V_0$ . Furthermore, it is likely that the approximation could be improved: by keeping  $\mathfrak{G}_2$  but ignoring correlations in higher-order GFs and calculating perturbatively further terms in  $V_0$  in operators  $\hat{m}$ .

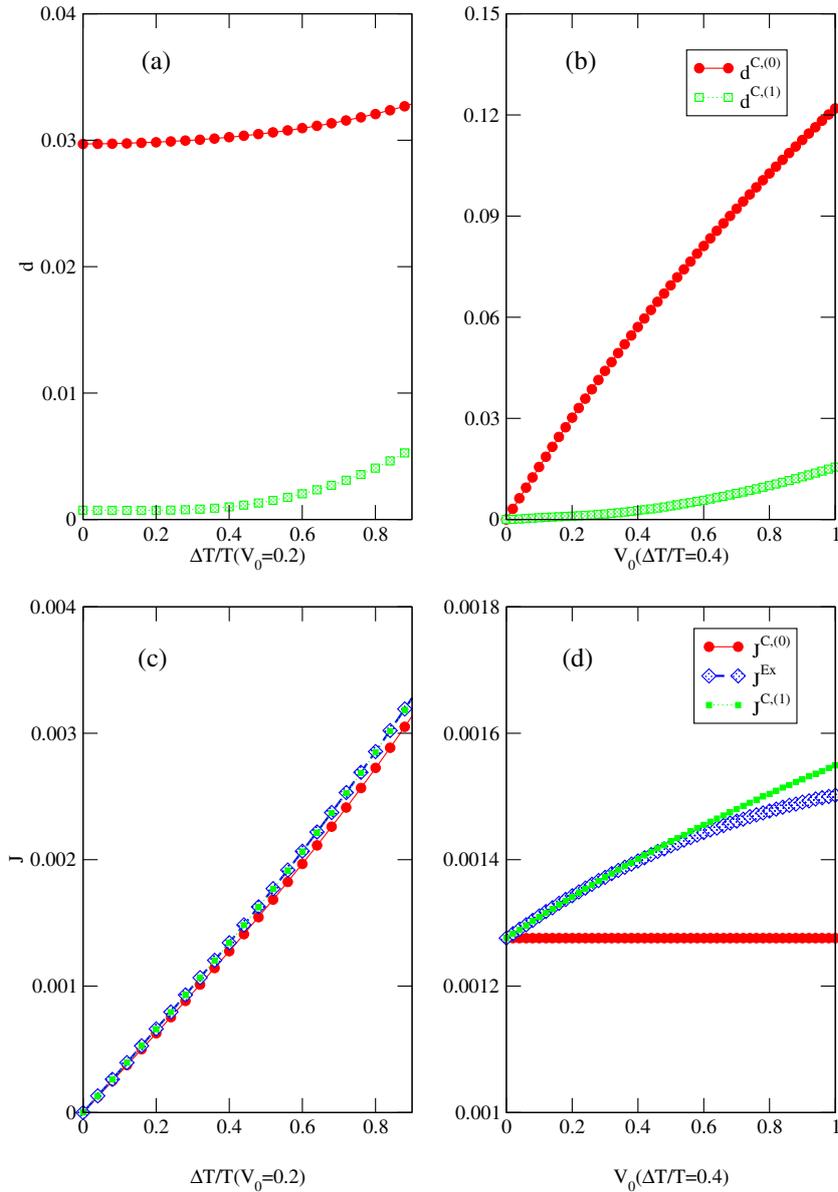
In order to estimate the accuracy of this approximation, let us assume that  $\lambda^2$  and  $V_0$  are small. Define similarly  $\Delta_n^{C,(0)} = g_n^{C,(0)} - g_n^{\text{Ex}}$  and  $\Delta_n^{C,(1)} = g_n^{C,(1)} - g_n^{\text{Ex}}$ . Again, we start from the equations of the three,  $g_1^{C,(0)}$ ,  $g_1^{C,(1)}$  and  $g_1^{\text{Ex}}$ , and then compare the three equations while ignoring certain higher-order terms, such as terms that are proportional to  $\lambda^2 V_0$ . See appendix C for more details of those equations and the estimation. We arrive at

$$\Delta_1^{C,(0)} = -i V_0 (\Gamma_0^{(1)})^{-1} g_2^{\text{Ex}} \sim V_0 |g_1^{\text{Ex}}|^2 \quad (31)$$

and

$$\Delta_1^{C,(1)} = (\Gamma_0^{(1)})^{-1} [i V_0^2 (\Gamma_0^{(2)})^{-1} g_3^{\text{Ex}} + \lambda^2 V_0 (\Gamma_0^{(2)})^{-1} \Delta_1^{C,(0)}] \sim V_0^2 |g_1^{\text{Ex}}|^3 + \lambda^2 V_0^2 |g_1^{\text{Ex}}|^2. \quad (32)$$

This agrees with the numerical tests that  $\Delta_1^{C,(0)}$  is proportional to  $V_0$ , while  $\Delta_1^{C,(1)}$  is proportional to  $V_0^2$ . We refer the readers to appendix C for definitions of all  $\Gamma$  matrices. Most importantly, we see again that  $\Delta_1^{C,(0)}$  is multiplied by a small number  $\lambda^2 V_0$  and then becomes a part of  $\Delta_1^{C,(1)}$ . The other term,  $V_0^2 g_3^{\text{Ex}} \sim V_0^2 |g_1^{\text{Ex}}|^3$ , since roughly  $|g_n^{\text{Ex}}| = |g^{\text{Ex}}|^n$ , is also much smaller than  $\Delta_1^{C,(0)} \sim V_0 |g_1^{\text{Ex}}|^2$ . However, for large enough  $V_0$ , the other approximation used in this method, the perturbation expansion of operators  $\hat{m}$ , will be invalid. Therefore, as long as  $V_0$  is a



**Figure 2.**  $g_1^{C,(1)}$  is compared with  $g_1^{Ex}$  for non-equilibrium interacting systems. (a)  $V = 0.2$ , accuracy was checked with different values of  $\Delta T$ . (b)  $\Delta T = 0.4T$ , accuracy was checked with different values of  $V_0$ . In both cases,  $d^{C,(1)}$  is always much smaller than  $d^{C,(0)}$ .  $J^{C,(0)}$  and  $J^{C,(1)}$  are compared against  $J^{Ex}$  in (c) and (d). From (c) we see that for a given value of  $V_0$ , as long as  $V_0$  is not too large,  $J^{C,(0)}$  already provides a major contribution. From (d) we find that for a relatively larger  $V_0$ , the difference between  $J^{C,(1)}$  and  $J^{C,(0)}$  becomes more important. At the same time, the difference between  $J^{C,(1)}$  and  $J^{Ex}$  also becomes larger for larger  $V_0$ .

small number compared with  $t$ , the method under consideration is very reasonable. It should be noted that this method is capable of dealing with large systems since it does not involve a direct diagonalization of a  $2^N$ -dimension matrix.

#### 4. Conclusion and discussion

To conclude, a BBGKY-like equation hierarchy is derived from the Redfield equation and two systematic approximations are suggested to solve the hierarchy. Using the first-order form of the two methods, NESSs of interacting systems are calculated. It is found that they are consistent with results made available by other direct methods. We also estimate the accuracy of the two approximations. The difference between the two is also discussed, showing that the first method is applicable for large  $V_0$  whereas the second method is more efficient, so it can be applied to larger systems. We have not tested the performance of further orders of both methods, although it seems quite straightforward. Our method can also be applied to the local-operator Lindblad equation [15, 20]. Also, it may be worth pursuing a comparison between results from the Lindblad equation and from the Redfield equation. Besides their applicability to NESSs, these methods may also be valuable for perturbation theory on equilibrium states. There, using the second method, the equilibrium interacting  $g_1^{C,(1)}$  can be calculated starting from the equilibrium non-interacting  $g_1^{C,(0)}$  by setting  $T_L = T_R$  and  $\mu_L = \mu_R$ . The computational cost, being a sequence of linear systems with dimension  $N^2$ , is obviously cheaper than direct diagonalization. It will be interesting to see a further comparison of accuracy and efficiency between these methods and other perturbative methods on equilibrium states. The non-equilibrium equal-time GFs calculated by the proposed methods are also objects of the NEGF method. Investigating the relation between these methods and the NEGF method may also be interesting.

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#### Appendix A. Perturbation decomposition of $\hat{m}$

From its definition in equation (7), in the representation of eigenvalues  $\{E_m\}$  and eigenstates  $\{|m\rangle\}$  of  $H_S$ , operator  $\hat{m}$  can be written as

$$\begin{aligned} (\hat{m}_\alpha)_{mn} &= (c_\alpha)_{mn} \sum_k |V_k^\alpha|^2 \int_0^\infty d\tau e^{i(E_n - E_m - \omega_{k,\alpha})\tau} \langle 1 - n_\alpha(\omega_{k,\alpha}) \rangle \\ &= (c_\alpha)_{mn} \pi \int d\omega D^\alpha(\omega) |V^\alpha(\omega)|^2 \langle 1 - n_\alpha(\omega) \rangle \delta(\Omega_{mn} - \omega) \\ &= (c_\alpha)_{mn} \pi D^\alpha(E_n - E_m) |V^\alpha(E_n - E_m)|^2 \langle 1 - n_\alpha(E_n - E_m) \rangle, \end{aligned} \quad (\text{A.1})$$

where we have used  $\int_0^\infty d\tau e^{i\omega\tau} = \pi\delta(\omega) + iP(\frac{1}{\omega})$  and neglected the principal value part. We have also assumed that it is possible to perform a change of variable on  $V_k^\alpha$  such that it becomes  $V^\alpha(k_{nm})$ , where  $k_{nm}$  is defined by  $\omega_{k_{nm},\alpha} = \Omega_{nm}$ , i.e. a bath mode resonant with this transition. This limits the possible forms of  $V_k^\alpha$  and  $\omega_{k,\alpha}$ . For example, for a given energy  $\Omega_{nm}$ , there should be a unique value of  $V_{k_{nm}}^\alpha$ . In this work, we take  $V_k^\alpha$  as a constant so this condition is satisfied.  $D_\alpha(\omega)$  is the bath's density of states. We arrived at

$$\hat{m}_\alpha = \pi \sum_{m,n} |m\rangle \langle n| \langle m|c_\alpha|n\rangle (1 - n_\alpha(\Omega_{nm})) D_\alpha(\Omega_{nm}) |V_{k_{nm}}^\alpha|^2, \quad (\text{A.2a})$$

$$\hat{m}_\alpha = \pi \sum_{m,n} |m\rangle \langle n| \langle m| c_\alpha^\dagger |n\rangle n_\alpha(\Omega_{mn}) D_\alpha(\Omega_{mn}) |V_{k_{mn}}^\alpha|^2, \quad (\text{A.2b})$$

where  $\Omega_{mn} = E_m - E_n = -\Omega_{nm}$ . We furthermore set  $V_{k_{nm}}^\alpha D_\alpha(\omega)$  as a constant and absorb it into  $\lambda^2$ . This procedure involves a direct diagonalization of the isolated system  $H_S$ . One can avoid this by finding such operators  $\hat{m}$  perturbatively.

Next, assuming  $V_0$  is small, we want to express operator  $\hat{m}_\alpha$  in terms of  $\{c_m\}$  and  $V_0$ . When  $V_0 = 0$ , the system is a tight-binding open chain, the following basis transformation

$$c_k = \frac{1}{\sqrt{N}} \sum_{l=1}^N \sin \frac{kl\pi}{N+1} c_l \quad (\text{A.3})$$

diagonalizes  $H_0$ ,

$$H_0 = \sum_{k=1}^N \epsilon_k c_k^\dagger c_k, \quad (\text{A.4})$$

where

$$\epsilon_k = -2t \cos \frac{\pi k}{N+1}. \quad (\text{A.5})$$

Therefore,  $c_\alpha(t)$  is a linear function of all  $c_m$ ,

$$c_l^{(0)}(t) = \frac{2}{N+1} \sum_{km} \sin \frac{\pi kl}{N+1} \sin \frac{\pi km}{N+1} e^{-i\epsilon_k t} c_m. \quad (\text{A.6})$$

Hence,  $\hat{m}_\alpha$  is also a linear combination of all  $c_m$ s. One can imagine that for small  $V_0$ ,  $\hat{m}_L$  should not be too far from a linear combination. Denote  $c_l(t)$  when  $V_0 = 0$  as  $c_l^{(0)}(t)$ . Starting from treating this as the zeroth order to the full dynamical  $c_l(t)$ , and expanding

$$c_l(t) = \sum_n V_0^n c_l^{(n)}(t), \quad (\text{A.7})$$

we may derive a perturbative equation of  $c_l^{(n)}(t)$ ,

$$\dot{c}_l^{(n)} = it(c_{l-1}^{(n)} + c_{l+1}^{(n)}) - id_l^{(n-1)}, \quad (\text{A.8})$$

where the shorthand notation, for non-negative integers  $n, n_1, n_2$  and  $n_3$ ,

$$d_l^{(n)} = \sum_{\substack{n_1, n_2, n_3 \\ \sum_i n_i}} \{c_l^{(n_1)} c_{l-1}^{\dagger, (n_2)} c_{l-1}^{(n_3)} + c_l^{(n_1)} c_{l+1}^{\dagger, (n_2)} c_{l+1}^{(n_3)}\}. \quad (\text{A.9})$$

Then the solution of the above equation can be written as

$$c_l^{(n)}(t) = -i \int_0^t d\tau \frac{2}{N+1} \sum_{km} \sin \frac{\pi kl}{N+1} \sin \frac{\pi km}{N+1} e^{-i\epsilon_k(t-\tau)} d_m^{(n-1)}(\tau). \quad (\text{A.10})$$

Here the initial condition that  $c^{(n)}(0) = 0$  ( $\forall n \geq 1$ ) is used. Plugging this general solution into equation (7), and after straightforward but tedious algebra, we arrive at the decomposition of  $\hat{m}_\alpha$  and  $\hat{m}_\alpha$  in equation (26) with expansion coefficients defined as

$$\mathcal{D}_{\alpha; m} = \pi \frac{2}{N+1} \sum_k \sin \frac{\pi kl_\alpha}{N+1} \sin \frac{\pi km}{N+1} [1 - n(\epsilon_k, T_\alpha)], \quad (\text{A.11a})$$

$$\bar{\mathcal{D}}_{\alpha;m} = \pi \frac{2}{N+1} \sum_k \sin \frac{\pi k l_\alpha}{N+1} \sin \frac{\pi k m}{N+1} n(\epsilon_k, T_\alpha), \quad (\text{A.11b})$$

$$\begin{aligned} \mathcal{D}_{\alpha;m_1 m_2 m_3} = & \pi \sum_{k,m,k_1,k_2,k_3} \left( \frac{2}{N+1} \right)^4 \frac{n(T_\alpha, \epsilon(k)) - n(T_\alpha, \epsilon(k_1) + \epsilon(k_3) - \epsilon(k_2))}{\epsilon(k_1) + \epsilon(k_3) - \epsilon(k_2) - \epsilon(k)} \\ & \times \sin \frac{k\pi l_\alpha}{N+1} \sin \frac{k_1\pi m_1}{N+1} \sin \frac{k_2\pi m_2}{N+1} \sin \frac{k_3\pi m_3}{N+1} \sin \frac{k\pi m}{N+1} \sin \frac{k_1\pi m}{N+1} \\ & \times \left( \sin \frac{k_2\pi(m+1)}{N+1} \sin \frac{k_3\pi(m+1)}{N+1} + \sin \frac{k_2\pi(m-1)}{N+1} \sin \frac{k_3\pi(m-1)}{N+1} \right). \end{aligned} \quad (\text{A.11c})$$

## Appendix B. Proof of non-equilibrium Wick's theorem

In this section, we will prove that when  $V_0 = 0$ ,<sup>1</sup>

$$G_2(k_1^\dagger, k_2^\dagger, k_3, k_4) = G_1(k_1^\dagger, k_4)G_1(k_2^\dagger, k_3) - G_1(k_1^\dagger, k_3)G_1(k_2^\dagger, k_4). \quad (\text{B.1})$$

Here, working in the momentum representation, defined in equations (A.3)–(A.5), is more convenient than the position representation. Starting from equation (8) with  $H_0$  in momentum space defined in equation (A.4) and using  $A = c_{k_1}^\dagger c_{k_2}$  and  $A = c_{k_1}^\dagger c_{k_2}^\dagger c_{k_3} c_{k_4}$ , we have the equations of, respectively,  $G_1(k_1^\dagger, k_2)$  and  $G_2(k_1^\dagger, k_2^\dagger, k_3, k_4)$  as follows,

$$\begin{aligned} 0 = & i(\epsilon_{k_2} - \epsilon_{k_1})G_1(k_1^\dagger, k_2) - \lambda^2 \frac{2\pi}{N+1} \sum_\alpha \sin \frac{k_1\pi l_\alpha}{N+1} \sin \frac{k_2\pi l_\alpha}{N+1} (n(k_1) + n(k_2)) \\ & + \lambda^2 \frac{2\pi}{N+1} \sum_{\alpha,k} \left[ \sin \frac{k_2\pi l_\alpha}{N+1} \sin \frac{k\pi l_\alpha}{N+1} G_1(k_1^\dagger, k) + \sin \frac{k_1\pi l_\alpha}{N+1} \sin \frac{k\pi l_\alpha}{N+1} G_1(k^\dagger, k_2) \right] \end{aligned} \quad (\text{B.2a})$$

$$\begin{aligned} 0 = & i(\epsilon_{k_4} + \epsilon_{k_3} - \epsilon_{k_2} - \epsilon_{k_1})G_2(k_1^\dagger, k_2^\dagger, k_3, k_4) \\ & + \lambda^2 \frac{2\pi}{N+1} \sum_{\alpha,k} \sin \frac{k_1\pi l_\alpha}{N+1} \sin \frac{k\pi l_\alpha}{N+1} G_2(k^\dagger, k_2^\dagger, k_3, k_4) \\ & + \lambda^2 \frac{2\pi}{N+1} \sum_{\alpha,k} \sin \frac{k_2\pi l_\alpha}{N+1} \sin \frac{k\pi l_\alpha}{N+1} G_2(k_1^\dagger, k^\dagger, k_3, k_4) \\ & + \lambda^2 \frac{2\pi}{N+1} \sum_{\alpha,k} \sin \frac{k_3\pi l_\alpha}{N+1} \sin \frac{k\pi l_\alpha}{N+1} G_2(k_1^\dagger, k_2^\dagger, k, k_4) \\ & + \lambda^2 \frac{2\pi}{N+1} \sum_{\alpha,k} \sin \frac{k_4\pi l_\alpha}{N+1} \sin \frac{k\pi l_\alpha}{N+1} G_2(k_1^\dagger, k_2^\dagger, k_3, k) \end{aligned}$$

<sup>1</sup> In a general non-equilibrium state, one may have  $\langle C_{k_3} C_{k_4} \rangle \neq 0$ . In that case, the above Wick's theorem should have a more general form and thus the cluster expansion will also have a more general form. In this work, our choice of coupling makes such GFs be equal to zero.

$$\begin{aligned}
& +\lambda^2 \frac{2\pi}{N+1} \sum_{\alpha} \sin \frac{k_2\pi l_{\alpha}}{N+1} \sin \frac{k_4\pi l_{\alpha}}{N+1} G_1(k_1^{\dagger}, k_3)(n(k_2) + n(k_4)) \\
& -\lambda^2 \frac{2\pi}{N+1} \sum_{\alpha} \sin \frac{k_2\pi l_{\alpha}}{N+1} \sin \frac{k_3\pi l_{\alpha}}{N+1} G_1(k_1^{\dagger}, k_4)(n(k_2) + n(k_3)) \\
& -\lambda^2 \frac{2\pi}{N+1} \sum_{\alpha} \sin \frac{k_1\pi l_{\alpha}}{N+1} \sin \frac{k_4\pi l_{\alpha}}{N+1} G_1(k_2^{\dagger}, k_3)(n(k_1) + n(k_4)) \\
& +\lambda^2 \frac{2\pi}{N+1} \sum_{\alpha} \sin \frac{k_1\pi l_{\alpha}}{N+1} \sin \frac{k_3\pi l_{\alpha}}{N+1} G_1(k_2^{\dagger}, k_4)(n(k_1) + n(k_3)). \tag{B.2b}
\end{aligned}$$

The combination of the two is a closed linear equation and has a unique solution. Therefore, we only need to find one solution. We first apply equations (B.1) to (B.2b) to expand  $G_2$  into products of  $G_1$ . It is then easy to prove that the resulting equation is equivalent to equation (B.2a), meaning that a solution of equation (B.2a) is also a solution of equation (B.2b). For example, if we collect terms with  $G_1(k_2^{\dagger}, k_4)$  together, we will have

$$\begin{aligned}
G_1(k_2^{\dagger}, k_4) \left\{ i(\epsilon(k_3) - \epsilon(k_1))G_1(k_1^{\dagger}, k_3) - \lambda^2 \frac{2\pi}{N+1} \sum_{\alpha} \sin \frac{k_1\pi l_{\alpha}}{N+1} \sin \frac{k_3\pi l_{\alpha}}{N+1} (n(k_1) + n(k_3)) \right. \\
\left. + \lambda^2 \frac{2\pi}{N+1} \sum_{\alpha, k} \left[ \sin \frac{k_3\pi l_{\alpha}}{N+1} \sin \frac{k\pi l_{\alpha}}{N+1} G_1(k_1^{\dagger}, k) + \sin \frac{k_1\pi l_{\alpha}}{N+1} \sin \frac{k\pi l_{\alpha}}{N+1} G_1(k^{\dagger}, k_3) \right] \right\}, \tag{B.3}
\end{aligned}$$

where the term in curly brackets is zero according to equation (B.2a). Therefore, solutions from equation (B.2a) also satisfy equation (B.2b), as long as equation (B.1), the non-equilibrium Wick's theorem, holds. Because of the uniqueness, the above solution is the only solution and thus equation (B.1) has to be satisfied.

### Appendix C. Estimation of convergence

In this section, we present our estimation of the leading order of  $G_1$ , such as  $\Delta_1^{E,(1)}$  and  $\Delta_1^{C,(1)}$ . We will see that  $\Delta_1^{E,(1)}$  in fact involves  $\Delta_2^{E,(0)}$ , which in turn needs the equation of  $G_2$ , the second equation of the hierarchy, derived using  $A = c_m^{\dagger} c_n^{\dagger} c_{m'} c_{n'}$ . A similar equation is needed for estimation of  $\Delta_1^{C,(1)}$ .

#### C.1. On $\Delta_1^{E,(1)}$ from method 1

After dropping the  $g_D$  term and the term that is proportional to  $\lambda^2 \Delta T$ , and keeping only up to the linear order of  $\Delta T$ ,  $g_1^{\text{Ex}}$ ,  $g_1^{E,(0)}$  and  $g_1^{E,(1)}$  respectively satisfy

$$(\Gamma_0^{(1)} + \Gamma_{,T}^{(1)} \Delta T) g_1^{\text{Ex}} = iV_0 g_2^{\text{Ex}} + \lambda^2 \nu_0, \tag{C.1a}$$

$$\Gamma_0^{(1)} g_1^{E,(0)} = iV_0 g_2^{E,(0)} + \lambda^2 \nu_0, \tag{C.1b}$$

$$(\Gamma_0^{(1)} + \Gamma_{,T}^{(1)} \Delta T) g_1^{E,(1)} = iV_0 g_2^{E,(0)} + \lambda^2 \nu_0, \tag{C.1c}$$

where  $\Gamma_0^{(1)} + \Gamma_T^{(1)} \Delta T$  is the zeroth and first order in  $\Delta T$  from  $\Gamma^{(1)}$  of equation (18).  $\Gamma_T^{(1)}$  denotes formally a derivative of  $T$  on  $\Gamma^{(1)}$ . To consider  $\Delta_1^{E,(0)}$ , one may use equation (C.1a) and equation (C.1b),

$$\Delta_1^{E,(0)} = \Delta T (\Gamma_0^{(1)})^{-1} \Gamma_T^{(1)} g_1^{\text{Ex}} + iV_0 (\Gamma_0^{(1)})^{-1} \Delta_2^{E,(0)}. \quad (\text{C.2})$$

$\Delta_1^{E,(1)}$  can be estimated from equation (C.1a) and (C.1c),

$$\Delta_1^{E,(1)} = iV_0 (\Gamma_0^{(1)} + \Gamma_T^{(1)} \Delta T)^{-1} \Delta_2^{E,(0)}, \quad (\text{C.3})$$

where  $\Delta_2^{E,(0)}$  is required. We find that, roughly speaking,  $\Delta_1^{E,(1)}$  takes the second term of  $\Delta_1^{E,(0)}$  but drops the first term. Therefore, next we only need to show that the second,  $iV_0 (\Gamma_0^{(1)})^{-1} \Delta_2^{E,(0)}$ , is much smaller than the first, or equivalently, smaller than the whole  $\Delta_1^{E,(0)}$ .

Estimation of  $\Delta_2^{E,(0)}$  involves the second equation of the hierarchy, i.e. the equation of  $G_2$ , which can be derived from substituting equation (12), the expression of operators  $\hat{m}$ , into equation (10),

$$\begin{aligned} 0 = & i\text{t} \langle c_m^\dagger c_n^\dagger c_{m'} c_{n'+1} \rangle + i\text{t} \langle c_m^\dagger c_n^\dagger c_{m'} c_{n'-1} \rangle + i\text{t} \langle c_m^\dagger c_n^\dagger c_{m'+1} c_{n'} \rangle + i\text{t} \langle c_m^\dagger c_n^\dagger c_{m'-1} c_{n'} \rangle \\ & - i\text{t} \langle c_m^\dagger c_{n-1}^\dagger c_{m'} c_{n'} \rangle - i\text{t} \langle c_m^\dagger c_{n+1}^\dagger c_{m'} c_{n'} \rangle - i\text{t} \langle c_{m-1}^\dagger c_n^\dagger c_{m'} c_{n'} \rangle - i\text{t} \langle c_{m+1}^\dagger c_n^\dagger c_{m'} c_{n'} \rangle \\ & + \lambda^2 \sum_{l,\alpha} \langle \delta_{n'\alpha} \bar{d}_{\alpha;l} c_m^\dagger c_n^\dagger c_{m'} c_l + \delta_{m'\alpha} \bar{d}_{\alpha;l} c_m^\dagger c_n^\dagger c_l c_{n'} + \delta_{n\alpha} \bar{d}_{\alpha;l} c_m^\dagger c_l c_{m'} c_{n'} + \delta_{m\alpha} \bar{d}_{\alpha;l} c_l c_n^\dagger c_{m'} c_{n'} \rangle \\ & + \lambda^2 \sum_{l,\alpha} \langle \delta_{n\alpha} \bar{d}_{\alpha;l}^* c_m^\dagger c_l c_{m'} c_{n'} + \delta_{m\alpha} \bar{d}_{\alpha;l}^* c_l c_n^\dagger c_{m'} c_{n'} + \delta_{n'\alpha} \bar{d}_{\alpha;l}^* c_m^\dagger c_n^\dagger c_{m'} c_l + \delta_{m'\alpha} \bar{d}_{\alpha;l}^* c_m^\dagger c_n^\dagger c_l c_{n'} \rangle \\ & + iV_0 \langle c_m^\dagger c_n^\dagger c_{m'} c_{n'} \rangle (\delta_{m'+1,n'} + \delta_{m'-1,n'} - \delta_{m+1,n} - \delta_{m-1,n}) \end{aligned} \quad (\text{C.4a})$$

$$-iV_0 \sum_{l=m\pm 1, n\pm 1} \langle c_l^\dagger c_m^\dagger c_n^\dagger c_l c_{m'} c_{n'} \rangle + iV_0 \sum_{l=m'\pm 1, n'\pm 1} \langle c_l^\dagger c_m^\dagger c_n^\dagger c_l c_{m'} c_{n'} \rangle \quad (\text{C.4b})$$

$$\begin{aligned} & -\lambda^2 \sum_{\alpha} [\delta_{n\alpha} \bar{d}_{\alpha;m'} \langle c_m^\dagger c_{n'} \rangle + \delta_{m\alpha} \bar{d}_{\alpha;n'} \langle c_n^\dagger c_{m'} \rangle + \delta_{m'\alpha} \bar{d}_{\alpha;n}^* \langle c_m^\dagger c_{n'} \rangle + \delta_{n'\alpha} \bar{d}_{\alpha;m}^* \langle c_n^\dagger c_{m'} \rangle] \\ & + \lambda^2 \sum_{\alpha} [\delta_{n\alpha} \bar{d}_{\alpha;n'} \langle c_m^\dagger c_{m'} \rangle + \delta_{m\alpha} \bar{d}_{\alpha;m'} \langle c_n^\dagger c_{n'} \rangle + \delta_{n'\alpha} \bar{d}_{\alpha;n}^* \langle c_m^\dagger c_{m'} \rangle + \delta_{m'\alpha} \bar{d}_{\alpha;m}^* \langle c_n^\dagger c_{n'} \rangle] \end{aligned} \quad (\text{C.4c})$$

$$\begin{aligned} & -\lambda^2 V_0 \sum_{\alpha} \langle \delta_{m'\alpha} c_m^\dagger c_n^\dagger c_{n'} D_{\alpha} - \delta_{n'\alpha} c_m^\dagger c_n^\dagger c_{m'} D_{\alpha} + \delta_{m\alpha} c_n^\dagger c_{m'} c_{n'} \bar{D}_{\alpha} - \delta_{n\alpha} c_m^\dagger c_{m'} c_{n'} \bar{D}_{\alpha} \rangle \\ & -\lambda^2 V_0 \sum_{\alpha} \langle \delta_{n'\alpha} \bar{D}_{\alpha}^{\dagger} c_m^\dagger c_n^\dagger c_{m'} - \delta_{m'\alpha} \bar{D}_{\alpha}^{\dagger} c_m^\dagger c_n^\dagger c_{n'} + \delta_{n\alpha} D_{\alpha}^{\dagger} c_m^\dagger c_{m'} c_{n'} - \delta_{m\alpha} D_{\alpha}^{\dagger} c_n^\dagger c_{m'} c_{n'} \rangle, \end{aligned} \quad (\text{C.4d})$$

which will be denoted in the following compactly as

$$\Gamma^{(2)} g_2^{\text{Ex}} = iV_0 g_3^{\text{Ex}} + \lambda^2 g_1^{\text{Ex}} + \lambda^2 V_0 g_{D3}^{\text{Ex}}, \quad (\text{C.5})$$

where the vector  $g_2^{\text{Ex}}$  is defined similarly to  $g_1^{\text{Ex}}$  as follows,

$$g_2^{\text{Ex}} = [G_2(1^\dagger, 1^\dagger, 1, 1), G_2(1^\dagger, 1^\dagger, 1, 2), \dots, G_2(N^\dagger, N^\dagger, N, N)]^T. \quad (\text{C.6})$$

Note that some of the elements of  $g_2^{\text{Ex}}$  are naturally zero, but we still include them in this vector.  $g_3^{\text{Ex}}$ ,  $g_1^{\text{Ex}}$  and  $g_{D3}^{\text{Ex}}$  come from ordering, respectively, equation (C.4b), (C.4c) and (C.4d) in the same way as  $g_2^{\text{Ex}}$ . The matrix  $\Gamma^{(2)}$  can be read off from equation (C.4a). For example, assuming that  $m, n, m'$  and  $n'$  are all different and not equal to 1 or  $N$ , for simplicity, we have

$$\Gamma_{mN^3+nN^2+m'N+n', mN^3+nN^2+m'N+n'+1}^{(2)} = it. \quad (\text{C.7})$$

Since  $\lambda^2 V_0 \ll V_0$ , we drop the last  $g_{D3}^{\text{Ex}}$  term in the following estimation of the accuracy. Therefore, the exact solution and the equilibrium solution satisfy, respectively,

$$(\Gamma_0^{(2)} + \Gamma_{,T}^{(2)} \Delta T) g_2^{\text{Ex}} = iV_0 g_3^{\text{Ex}} + \lambda^2 g_1^{\text{Ex}}, \quad (\text{C.8a})$$

$$\Gamma_0^{(2)} g_2^{E,(0)} = iV_0 g_3^{E,(0)} + \lambda^2 g_1^{E,(0)}, \quad (\text{C.8b})$$

where  $\Gamma_0^{(2)}$  and  $\Gamma_{,T}^{(2)} \Delta T$  stand for the zeroth and first order in  $\Delta T$  in  $\Gamma^{(2)}$ . Now  $\Delta_2^{E,(0)}$  can be analyzed,

$$\Delta_2^{E,(0)} = iV_0 (\Gamma_0^{(2)})^{-1} \Delta_3^{E,(0)} + (\Gamma_0^{(2)})^{-1} \Gamma_{,T}^{(2)} \Delta T g_2^{\text{Ex}} + (\Gamma_0^{(2)})^{-1} \lambda^2 \Delta_1^{E,(0)}. \quad (\text{C.9})$$

Here in fact  $\Gamma^{(1)}$  and  $\Gamma^{(2)}$  have different dimensions. However, in this estimation of the order of magnitudes, we ignore this difference and, furthermore, the matrices are regarded as constants with order 1. For the moment, let us focus on the last term of  $\Delta_2^{E,(0)}$ . Recall that we want to compare  $iV_0 (\Gamma_0^{(1)})^{-1} \Delta_2^{E,(0)}$  against  $\Delta_1^{E,(0)}$ . Focusing only on the last term, we have

$$iV_0 \Delta_2^{E,(0)} \approx iV_0 \lambda^2 \Delta_1^{E,(0)}, \quad (\text{C.10})$$

which is much smaller than  $\Delta_1^{E,(0)}$  as long as  $V_0 \lambda^2 \ll 1$ . The effect of the other two terms has been discussed in the main text in section 3.1.1.

## C.2. On $\Delta_1^{C,(1)}$ from method 2

In this case, after dropping the  $\Gamma_D^{(1)}$  term and terms that are proportional to  $\lambda^2 V_0$ ,  $g_1^{\text{Ex}}$ ,  $g_1^{C,0}$  and  $g_1^{C,(1)}$ , respectively, satisfy the following equations,

$$\Gamma_0^{(1)} g_1^{\text{Ex}} = iV_0 g_2^{\text{Ex}} + \lambda^2 \nu_0, \quad (\text{C.11a})$$

$$\Gamma_0^{(1)} g_1^{C,(0)} = \lambda^2 \nu_0, \quad (\text{C.11b})$$

$$\Gamma_0^{(1)} g_1^{C,(1)} = iV_0 g_2^{C,(1)} + \lambda^2 \nu_0. \quad (\text{C.11c})$$

From equation (C.11b) and (C.11a), one may find

$$\Delta_1^{C,(0)} = -iV_0 (\Gamma_0^{(1)})^{-1} g_2^{\text{Ex}}. \quad (\text{C.12})$$

Comparing equation (C.11c) and equation (C.11a), one obtains

$$\Delta_1^{C,(1)} = iV_0 (\Gamma_0^{(1)})^{-1} (g_2^{C,(1)} - g_2^{\text{Ex}}). \quad (\text{C.13})$$

Note that the magnitude of  $\Delta_2^{C,(1)} = (g_2^{C,(1)} - g_2^{\text{Ex}})$  is in fact smaller than the magnitude of  $\Delta_2^{C,(0)} = (g_2^{C,(0)} - g_2^{\text{Ex}})$ , which involves the second equation of the hierarchy, i.e. the equation of  $G_2$ . So we may analyze the latter to get an upper bound of the former. In this case, one needs to substitute equation (26) into equation (10). The resulting equation will have the same structure as equation (C.4) but every  $d_{\alpha;l}$  and  $\bar{d}_{\alpha;l}$  is replaced, respectively, by  $\mathcal{D}_{\alpha;m}$  and  $\bar{\mathcal{D}}_{\alpha;m}$ , and a similar substitution on  $D_\alpha$  and  $\bar{D}_\alpha$ . Ignoring terms that are proportional to  $\lambda^2 V_0$ ,  $g_2^{\text{Ex}}$  and  $g_2^{C,(0)}$  are, respectively, the solutions of

$$\Gamma_0^{(2)} g_2^{\text{Ex}} = iV_0 g_3^{\text{Ex}} + \lambda^2 g_1^{\text{Ex}}, \quad (\text{C.14a})$$

$$\Gamma_0^{(2)} g_2^{C,(0)} = \lambda^2 g_1^{C,(0)}. \quad (\text{C.14b})$$

Comparing these two equations, we find that

$$\Delta_2^{C,(0)} = -iV_0(\Gamma_0^{(2)})^{-1} g_3^{\text{Ex}} - \lambda^2(\Gamma_0^{(2)})^{-1} \Delta_1^{C,(0)}. \quad (\text{C.15})$$

Focusing only on the last term, we have

$$iV_0 \Delta_2^{C,(0)} \approx -iV_0 \lambda^2 \Delta_1^{C,(0)}, \quad (\text{C.16})$$

which is much smaller than  $\Delta_1^{C,(0)}$  as long as  $V_0 \lambda^2 \ll 1$ . The effect of the first term has been discussed in the main text in section 3.2.1.

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