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Time propagation of the Kadanoff–Baym equations for inhomogeneous systems

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We have developed a time-propagation scheme for the Kadanoff–Baym equations for general inhomogeneous systems. These equations describe the time evolution of the nonequilibrium Green function for interacting many-body systems in the presence of time-dependent external fields. The external fields are treated nonperturbatively whereas the many-body interactions are incorporated perturbatively using Φ -derivable self-energy approximations that guarantee the satisfaction of the macroscopic conservation laws of the system. These approximations are discussed in detail for the time-dependent Hartree–Fock, the second Born, and the *GW* approximation. © 2009 American Institute of Physics. [DOI: 10.1063/1.3127247]

I. INTRODUCTION

The recent developments in the field of molecular electronics have emphasized the need for further development of theoretical methods that allow for a systematic study of dynamical processes such as relaxation and decoherence at the nanoscale. Understanding these processes is of utmost importance for making progress in molecular electronics, whose ultimate goal is to minimize the size and maximize the speed of integrated devices.¹ To study these phenomena, theoretical methods must allow for the possibility to study the ultrafast transient dynamics^{2,3} up to the picosecond^{4,5} and femtosecond time scale, while including Coulomb interactions, without violating basic conservation laws such as the continuity equation.⁶ A theoretical framework that incorporates these features is the nonequilibrium Green function approach based on the real time propagation of the Kadanoff–Baym (KB) equations.^{7–14} This method allows for systematic inclusion of electron interactions while providing results in agreement with the macroscopic conservation laws of the system.^{6,8} In two recent letters^{7,11} we applied the KB equations to investigate the short time dynamics of atoms and molecules in time-dependent external fields, as well as the transport dynamics of double quantum dot devices. It is the aim of this paper to describe in detail the underlying method that was only briefly described in those letters. This includes both a description of the theory as well as the time-propagation algorithm. We further generalize the equilibrium method, described in two recent papers,^{15,16} to the nonequilibrium domain. We also extend earlier work on the time-propagation method of the KB equations for homogeneous systems^{17,18} to the case of inhomogeneous systems. In the inhomogeneous case we cannot take advantage of Fourier transform techniques anymore. The KB equations become time-dependent matrix equations instead, in which the matrices

are indexed by basis function indices. The time-stepping algorithm has to take into account the special double-time structure of the equations which are furthermore nonlinear, inhomogeneous, and non-Hermitian. Therefore, several standard time-propagation methods cannot be used. Our approach is different from the one presented in Refs. 17 and 18 by incorporating correlated initial states and the memory thereof, which is described in terms of Green functions with mixed real and imaginary time arguments. To simplify the time-stepping procedure, we make use of several symmetry relations of the Green function.

This paper is divided as follows: in Sec. II we present the KB equations and their symmetry properties. In Sec. III we discuss the conserving self-energy approximations that we use, and in Sec. IV we present the time-propagation method that we developed for systems described within a general basis set representation. Finally in Sec. V we present a summary and conclusions.

II. THEORY

We consider a many-body system that is initially in equilibrium at a temperature T and with a chemical potential μ . At an initial time t_0 the system is exposed to a time-dependent external field. This external field can, for instance, be a bias voltage in a quantum transport case, or a laser pulse. The field forces the system out of equilibrium and we aim to describe the time evolution of this nonequilibrium state. In second quantization the time-dependent Hamiltonian of the system reads (throughout this paper we use atomic units $\hbar = m = e = 1$)

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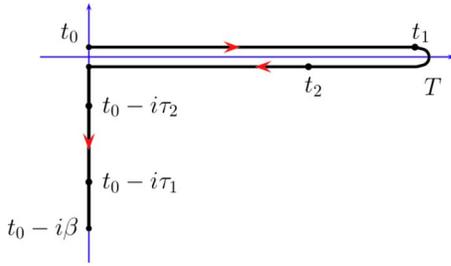


FIG. 1. (Color online) Keldysh contour. The depicted contour allows for the calculation of observables for times $t_0 \leq t \leq T$. The initial Green function is calculated on the imaginary track $[t_0, t_0 - i\beta]$. As we propagate the KB equations in time, for real times $t > t_0$, the turning point of the time contour at $t = T$ moves to the right along the real time axis.

$$\hat{H}(t) = \int d\mathbf{x} \hat{\psi}^\dagger(\mathbf{x}) h(\mathbf{x}, t) \hat{\psi}(\mathbf{x}) + \frac{1}{2} \int \int d\mathbf{x}_1 d\mathbf{x}_2 \hat{\psi}^\dagger(\mathbf{x}_1) \hat{\psi}^\dagger(\mathbf{x}_2) v(\mathbf{r}_1, \mathbf{r}_2) \hat{\psi}(\mathbf{x}_2) \hat{\psi}(\mathbf{x}_1), \quad (1)$$

where $\mathbf{x} = (\mathbf{r}, \sigma)$ denotes the space and spin coordinates. The two-body interaction will, in general, be a Coulombic repulsion of the form $v(\mathbf{r}_1, \mathbf{r}_2) = 1/|\mathbf{r}_1 - \mathbf{r}_2|$. The one-body part of the Hamiltonian is

$$h(\mathbf{x}, t) = -\frac{1}{2} \nabla^2 + w(\mathbf{x}, t) - \mu, \quad (2)$$

where $w(\mathbf{x}, t)$ is a time-dependent external potential. The chemical potential μ of the initial equilibrium system is absorbed in the one-body part of the Hamiltonian. The expectation value of an operator \hat{O} , for a system initially in thermodynamic equilibrium ($t < t_0$), is given by

$$\langle \hat{O} \rangle = \text{Tr}\{\hat{\rho} \hat{O}\}, \quad (3)$$

where $\hat{\rho} = e^{-\beta \hat{H}_0} / \text{Tr} e^{-\beta \hat{H}_0}$ is the statistical operator, \hat{H}_0 is the time-independent Hamiltonian that describes the system before the time-dependent field is applied and $\beta = 1/k_B T$ is the inverse temperature. The trace here represents a summation over a complete set of states in Fock space.¹⁹ After the time-dependent external field is switched on at time t_0 , the expectation value is given by

$$\langle \hat{O}(t) \rangle = \frac{\text{Tr}\{\hat{U}(t_0 - i\beta, t_0) \hat{O}_H(t)\}}{\text{Tr}\{\hat{U}(t_0 - i\beta, t_0)\}}, \quad (4)$$

where $\hat{O}_H(t) = \hat{U}(t_0, t) \hat{O} \hat{U}(t, t_0)$ is the operator \hat{O} in the Heisenberg picture and $\hat{U}(t_2, t_1) = T[\exp(-i \int_{t_1}^{t_2} dt \hat{H}(t))]$, for $t_2 > t_1$, is the time-ordered evolution operator of the system. We further wrote $\exp(-\beta \hat{H}_0) = \hat{U}(t_0 - i\beta, t_0)$ as an evolution operator in imaginary time. If we read the time arguments in Eq. (4) from right to left we see that they follow a time-contour as displayed in Fig. 1. This contour is also known as the Keldysh contour.^{20,21} A more detailed inspection of Eq. (4) then shows that the expectation value can also be written as a contour-ordered product.^{20,22–25} The one-particle Green function is then defined as a contour-ordered product of a creation and an annihilation operator

$$G(1, 2) = -i \langle T_C [\hat{\psi}_H(1) \hat{\psi}_H^\dagger(2)] \rangle, \quad (5)$$

where T_C denotes the time-ordering operator on the contour and where we used the compact notation $1 = (\mathbf{x}_1, t_1)$ and $2 = (\mathbf{x}_2, t_2)$. If we consider the Green function at time $t_1 = t_0 - i\beta$ and use the cyclic property of the trace, we find that $G(\mathbf{x}_1 t_0 - i\beta, 2) = -G(\mathbf{x}_1 t_0, 2)$.²⁴ Hence, the Green function defined in Eq. (5) obeys the boundary conditions

$$G(\mathbf{x}_1 t_0, 2) = -G(\mathbf{x}_1 t_0 - i\beta, 2), \quad (6)$$

$$G(1, \mathbf{x}_2 t_0) = -G(1, \mathbf{x}_2 t_0 - i\beta). \quad (7)$$

The Green function satisfies the equation of motion

$$[i\partial_{t_1} - h(1)]G(1, 2) = \delta(1, 2) + \int_C d3 \Sigma(1, 3)G(3, 2), \quad (8)$$

as well as a corresponding adjoint equation.^{9,24} In Eq. (8) the time integration is carried out along the contour C . The self-energy Σ incorporates the effects of exchange and correlation in many-particle systems and is a functional of the Green function that can be defined diagrammatically.^{9,19} The Green function can be written as

$$G(1, 2) = \theta(t, t') G^>(1, 2) + \theta(t', t) G^<(1, 2), \quad (9)$$

where θ is a step function generalized to arguments on the contour, i.e., with $\theta(t, t') = 1$ if t is later on the contour than t' and zero otherwise.⁹ The greater and lesser components $G^>$ and $G^<$ respectively, have the explicit form

$$G^>(1, 2) = -i \langle \hat{\psi}_H(1) \hat{\psi}_H^\dagger(2) \rangle, \quad (10)$$

$$G^<(1, 2) = i \langle \hat{\psi}_H^\dagger(2) \hat{\psi}_H(1) \rangle. \quad (11)$$

When one of the arguments is on the vertical track of the contour, we adopt the notation²²

$$G^l(1, \mathbf{x}_2, -i\tau_2) = G^<(1, \mathbf{x}_2, t_0 - i\tau_2), \quad (12)$$

$$G^f(\mathbf{x}_1, -i\tau_1, 2) = G^>(\mathbf{x}_1, t_0 - i\tau_1, 2). \quad (13)$$

Finally, for the case when both time arguments are on the imaginary track of the contour, we have the so-called Matsubara–Green function iG^M ,¹⁹

$$iG^M(\mathbf{x}_1 \tau_1, \mathbf{x}_2 \tau_2) = G(\mathbf{x}_1 t_0 - i\tau_1, \mathbf{x}_2 t_0 - i\tau_2), \quad (14)$$

which is a well-known object from the equilibrium theory. The factor i in the definition of Eq. (14) is a convention which ensures that G^M is a real function. The self-energy Σ has a similar general structure as the Green function

$$\Sigma(1, 2) = \Sigma^{\text{HF}}(1, 2) + \theta(t, t') \Sigma^>(1, 2) + \theta(t', t) \Sigma^<(1, 2). \quad (15)$$

The main difference with Eq. (9) is the appearance of the term Σ^{HF} which is proportional to a contour delta function $\delta(t_1, t_2)$ in the time coordinates.⁹ This term has the explicit form

$$\Sigma^{\text{HF}}[G](1, 2) = \delta(t_1, t_2) \Sigma^{\text{HF}}(\mathbf{x}_1, \mathbf{x}_2, t_1), \quad (16)$$

where

$$\begin{aligned} \Sigma^{\text{HF}}(\mathbf{x}_1, \mathbf{x}_2, t) &= iG^<(\mathbf{x}_1 t, \mathbf{x}_2 t)v(\mathbf{x}_1, \mathbf{x}_2) \\ &\quad - i\delta(\mathbf{x}_1 - \mathbf{x}_2) \int d\mathbf{x}_3 v(\mathbf{x}_1, \mathbf{x}_3)G^<(\mathbf{x}_3 t, \mathbf{x}_3 t). \end{aligned} \quad (17)$$

The structure of this self-energy is that of the Hartree–Fock (HF) approximation. However, in general, we will evaluate this expression for Green functions G obtained beyond HF level (see Sec. III). Using the form of the self-energy of Eq. (15) the contour integrations can be readily carried out^{9,20} and we find separate equations for the different Green functions G^{\lessgtr} , G^{\ll} and G^M . To display their temporal structure more clearly we suppress the spatial indices of the Green functions and self-energies. Alternatively, these quantities may be regarded as matrices.⁷ On the imaginary track of the contour we obtain

$$[-\partial_\tau - h]G^M(\tau) = \delta(\tau) + \int_0^\beta d\bar{\tau} \Sigma^M(\tau - \bar{\tau})G^M(\bar{\tau}), \quad (18)$$

where the Green function and the self-energy are functions of the time-differences only, i.e., $iG^M(\tau_1 - \tau_2) = G(-i\tau_1, -i\tau_2)$ and $i\Sigma^M(\tau_1 - \tau_2) = \Sigma(-i\tau_1, -i\tau_2)$, since the Hamiltonian is time independent (and equal to \hat{H}_0) on the imaginary track. Equation (18), which determines the Green function of the equilibrium system, has been treated in detail in Refs. 15 and 16. For the other Green functions we obtain

$$i\partial_t G^{\lessgtr}(t, t') = h^{\text{HF}}(t)G^{\lessgtr}(t, t') + I_1^{\lessgtr}(t, t'), \quad (19)$$

$$-i\partial_{t'} G^{\lessgtr}(t, t') = G^{\lessgtr}(t, t')h^{\text{HF}}(t') + I_2^{\lessgtr}(t, t'), \quad (20)$$

$$i\partial_t G^{\ll}(t, -i\tau) = h^{\text{HF}}(t)G^{\ll}(t, -i\tau) + I^{\ll}(t, -i\tau), \quad (21)$$

$$-i\partial_{t'} G^{\ll}(-i\tau, t) = G^{\ll}(-i\tau, t)h^{\text{HF}}(t) + I^{\ll}(-i\tau, t), \quad (22)$$

where $h^{\text{HF}}(t) = h(t) + \Sigma^{\text{HF}}(t)$ and $\Sigma^{\text{HF}}(t)$ is given by Eq. (17). The retarded and advanced functions for G and Σ are defined according to

$$F^{R/A}(t, t') = \pm \theta(\pm t \mp t') [F^>(t, t') - F^<(t, t')], \quad (23)$$

with F replaced by G and Σ , respectively. The so-called collision terms I^{\lessgtr} and I^{\ll} have the form

$$\begin{aligned} I_1^{\lessgtr}(t, t') &= \int_0^t d\bar{t} \Sigma^R(t, \bar{t})G^{\lessgtr}(\bar{t}, t') + \int_0^{t'} d\bar{t} \Sigma^{\lessgtr}(t, \bar{t})G^A(\bar{t}, t') \\ &\quad + \frac{1}{i} \int_0^\beta d\bar{\tau} \Sigma^{\ll}(t, -i\bar{\tau})G^{\ll}(-i\bar{\tau}, t'), \end{aligned} \quad (24)$$

$$\begin{aligned} I_2^{\lessgtr}(t, t') &= \int_0^t d\bar{t} G^R(t, \bar{t})\Sigma^{\lessgtr}(\bar{t}, t') + \int_0^{t'} d\bar{t} G^{\lessgtr}(t, \bar{t})\Sigma^A(\bar{t}, t') \\ &\quad + \frac{1}{i} \int_0^\beta d\bar{\tau} G^{\ll}(t, -i\bar{\tau})\Sigma^{\ll}(-i\bar{\tau}, t'), \end{aligned} \quad (25)$$

$$\begin{aligned} I^{\ll}(t, -i\tau) &= \int_0^t d\bar{t} \Sigma^R(t, \bar{t})G^{\ll}(\bar{t}, -i\tau) \\ &\quad + \int_0^\beta d\bar{\tau} \Sigma^{\ll}(t, -i\bar{\tau})G^M(\bar{\tau} - \tau), \end{aligned} \quad (26)$$

$$\begin{aligned} I^{\ll}(-i\tau, t) &= \int_0^t d\bar{t} G^{\ll}(-i\tau, \bar{t})\Sigma^A(\bar{t}, t) \\ &\quad + \int_0^\beta d\bar{\tau} G^M(\tau - \bar{\tau})\Sigma^{\ll}(-i\bar{\tau}, t). \end{aligned} \quad (27)$$

These equations are readily derived using the conversion table of Ref. 20. From the symmetry relations

$$G^{\lessgtr}(t, t')^\dagger = -G^{\lessgtr}(t', t), \quad (28)$$

$$\Sigma^{\lessgtr}(t, t')^\dagger = -\Sigma^{\lessgtr}(t', t), \quad (29)$$

it follows that we only need to calculate $G^>(t, t')$ and $\Sigma^>(t, t')$ for $t > t'$ and $G^<(t, t')$ and $\Sigma^<(t, t')$ for $t \leq t'$. These equations imply that $I_{1,2}^{\lessgtr}(t, t') = -I_{2,1}^{\lessgtr}(t', t)^\dagger$. We further have

$$G^{\ll}(-i\tau, t) = G^{\ll}(t, -i(\beta - \tau))^\dagger, \quad (30)$$

$$\Sigma^{\ll}(-i\tau, t) = \Sigma^{\ll}(t, -i(\beta - \tau))^\dagger. \quad (31)$$

The symmetry relations (28) and (30) for the Green function follow directly from its definition, whereas the symmetry relations (29) and (31) for the self-energy follow from Eqs. (3.19) and (3.20) of Ref. 9. Another consequence of Eqs. (30) and (31) is that $I^{\ll}(-i\tau, t) = [I^{\ll}(t, -i(\beta - \tau))]^\dagger$, which means that, in practice, it is sufficient to calculate only $I_1^>$, $I_2^<$, and I^{\ll} . Equations (19)–(22) are known as the Kadanoff–Baym equations.^{8,9}

Once the Matsubara–Green function $G^M(\tau)$ is obtained from Eq. (18), the Green functions $G^x(x = \lessgtr, \ll)$ can be calculated by time propagation. Their initial conditions are

$$G^>(0, 0) = iG^M(0^+), \quad (32)$$

$$G^<(0, 0) = iG^M(0^-), \quad (33)$$

$$G^{\ll}(0, -i\tau) = iG^M(-\tau), \quad (34)$$

$$G^{\ll}(-i\tau, 0) = iG^M(\tau). \quad (35)$$

The KB equations, together with the initial conditions, completely determine the Green functions for all times once a choice for the self-energy has been made. The form of the self-energy will be the topic of Sec. III.

III. SELF-ENERGY APPROXIMATIONS

In the applications of the KB equations it is possible to guarantee that the macroscopic conservation laws, such as those of particle, momentum, and energy conservation, are obeyed. Baym⁶ showed that this is the case whenever the self-energy is obtained from a functional $\Phi[G]$, such that

$$\Sigma(1,2) = \frac{\delta\Phi}{\delta G(2,1)}. \quad (36)$$

Such approximations to the self-energy are called conserving or Φ -derivable approximations. Well-known conserving approximations are the HF, the second Born,⁸ the GW ,²⁶ and the T -matrix⁸ approximation. In our work we implemented the first three of these.

The second Born approximation: This approximation for the self-energy consists of the two diagrams to second order in the two-particle interaction^{8,27}

$$\Sigma(1,2) = \Sigma^{\text{HF}}(1,2) + \Sigma^{(2)}(1,2), \quad (37)$$

where Σ^{HF} is the HF part of the self-energy of Eq. (16) and $\Sigma^{(2)} = \Sigma^{(2a)} + \Sigma^{(2b)}$ is the sum of the two terms

$$\begin{aligned} \Sigma^{(2a)}(1,2) = & -i^2 G(1,2) \int d3d4v(1,3) \\ & \times G(3,4)G(4,3)v(4,2), \end{aligned} \quad (38)$$

$$\begin{aligned} \Sigma^{(2b)}(1,2) = & i^2 \int d3d4G(1,3)v(1,4)G(3,4) \\ & \times G(4,2)v(3,2), \end{aligned} \quad (39)$$

where $v(1,2) = v(\mathbf{x}_1, \mathbf{x}_2)\delta(t_1, t_2)$. These terms are usually referred to as the second-order direct and exchange terms. This approximation to the self-energy has been discussed in detail for the equilibrium case in Ref. 15. For the nonequilibrium case we need to calculate the various components $\Sigma^x(x = \lessgtr, \lll)$. These are explicitly given by

$$\begin{aligned} \Sigma^{(2a),\lessgtr}(1,2) = & -i^2 G^{\lessgtr}(1,2) \int d3d4v(1,3) \\ & \times G^{\lessgtr}(3,4)G^{\gtrless}(4,3)v(4,2), \end{aligned} \quad (40)$$

$$\begin{aligned} \Sigma^{(2a),\lll}(1,2) = & -i^2 \int d3d4G^{\lll}(1,2)v(1,3) \\ & \times G^{\lll}(3,4)G^{\lll}(4,3)v(4,2), \end{aligned} \quad (41)$$

for the direct diagram, and

$$\begin{aligned} \Sigma^{(2b),\lessgtr}(1,2) = & i^2 \int d3d4G^{\lessgtr}(1,3)v(1,4)G^{\gtrless}(3,4) \\ & \times G^{\lessgtr}(4,2)v(3,2), \end{aligned} \quad (42)$$

$$\begin{aligned} \Sigma^{(2b),\lll}(1,2) = & i^2 \int d3d4G^{\lll}(1,3)v(1,4)G^{\lll}(3,4) \\ & \times G^{\lll}(4,2)v(3,2), \end{aligned} \quad (43)$$

for the second-order exchange diagram. These expressions follow immediately from Eqs. (38) and (39) with help of the conversion table of Ref. 20.

The GW approximation: In the GW approximation the exchange-correlation part of the self-energy is given as a product of the Green function G with a dynamically screened interaction W .²⁶ The screened interaction W satisfies the equation

$$W(1,2) = v(1,2) + \int d3d4v(1,3)P(3,4)W(4,2). \quad (44)$$

Here, v is the bare Coulomb interaction, and

$$P(1,2) = -iG(1,2)G(2,1), \quad (45)$$

is the irreducible polarization.²⁶ However, since the first term in Eq. (44) is singular in time (proportional to a delta function) it is convenient, for numerical purposes, to define its time-nonlocal part $\tilde{W} = W - v$.¹⁶ From Eq. (44) it follows that

$$\begin{aligned} \tilde{W}(1,2) = & \int d3d4v(1,3)P(3,4)v(4,2) \\ & + \int d3d4v(1,3)P(3,4)\tilde{W}(4,2). \end{aligned} \quad (46)$$

In terms of \tilde{W} , the self-energy has the form²⁶

$$\Sigma(1,2) = \Sigma^{\text{HF}}(1,2) + iG(1,2)\tilde{W}(1,2). \quad (47)$$

The part $\Sigma_c = iG\tilde{W}$ represents the correlation part of the self-energy and has the components

$$\Sigma_c^{\lessgtr}(1,2) = iG^{\lessgtr}(1,2)\tilde{W}^{\gtrless}(2,1), \quad (48)$$

$$\Sigma_c^{\lll}(1,2) = iG^{\lll}(1,2)\tilde{W}^{\lll}(2,1). \quad (49)$$

From the fact that $\tilde{W}(1,2)$ has the same symmetries as the contour-ordered density response function²⁶ $\chi(1,2) = -i\langle T_C[\hat{n}_H(1)\hat{n}_H(2)] \rangle$, where \hat{n} is the density operator, it follows that

$$\tilde{W}^{\lessgtr}(2,1) = \tilde{W}^{\gtrless}(1,2) = -[\tilde{W}^{\gtrless}(2,1)]^*, \quad (50)$$

$$W^{\lll}(1,2) = W^{\lll}(2,1). \quad (51)$$

In the following, we will again suppress the spatial coordinates in order to display the temporal structure of the equations more clearly. From the symmetry relations (50), (51), (48), and (49), and the fact that we only need $\Sigma^>(t, t')$ for $t > t'$ and $\Sigma^<(t, t')$ for $t \leq t'$, it follows that we only need to calculate $\tilde{W}^{\lll}(t, -i\tau)$, and $\tilde{W}^<(t, t')$ for $t \leq t'$. The latter obey the equations:

$$\tilde{W}^<(t, t') = vP^<(t, t')v + vX^<(t, t'), \quad (52)$$

$$\tilde{W}^{\lll}(t, -i\tau) = vP^{\lll}(t, -i\tau)v + vX^{\lll}(t, -i\tau), \quad (53)$$

where

$$P^<(t, t') = -iG^<(t, t')G^>(t', t), \quad (54)$$

$$P^{\lll}(t, -i\tau) = -iG^{\lll}(t, -i\tau)G^{\lll}(-i\tau, t), \quad (55)$$

and where the terms $X^<$ and X^{\lll} are given by

$$\begin{aligned} X^<(t, t') = & \int_0^{t'} d\bar{t}P^<(t, \bar{t})\tilde{W}^A(\bar{t}, t') + \int_0^t d\bar{t}P^R(t, \bar{t})\tilde{W}^<(\bar{t}, t') \\ & + \int_0^\beta d\bar{t}P^{\lll}(t, -i\bar{\tau})\tilde{W}^{\lll}(-i\bar{\tau}, t'), \end{aligned} \quad (56)$$

$$X^{\downarrow}(t, -i\tau) = \int_0^{t'} d\bar{t} P^R(t, \bar{t}) \tilde{W}^{\downarrow}(\bar{t}, -i\tau) + \int_0^{\beta} d\bar{\tau} P^{\downarrow}(t, -i\bar{\tau}) \tilde{W}^M(\bar{\tau} - \tau), \quad (57)$$

with the retarded and advanced quantities defined as in Eq. (23). The initial conditions for $\tilde{W}^<$ and \tilde{W}^{\downarrow} are

$$\tilde{W}^<(0, 0) = i\tilde{W}^M(0^-), \quad (58)$$

$$\tilde{W}^{\downarrow}(0, -i\tau) = i\tilde{W}^M(-\tau), \quad (59)$$

where $i\tilde{W}^M(\tau - \tau') = \tilde{W}(t_0 - i\tau, t_0 - i\tau')$ is the Matsubara interaction discussed in detail in Ref. 16.

IV. TIME PROPAGATION OF THE KADANOFF–BAYM EQUATIONS

In the following, we will describe the time-propagation method which we employed to solve the KB equations. This method can be applied to general Hamiltonians containing one- and two-body interactions, and is further independent of the explicit form of the self-energy.

The time-propagation method is applied to the KB equations in matrix form. This matrix form is obtained by expressing the Green function in terms of a set of basis functions $\phi_i(\mathbf{x})$, which we choose to be HF orbitals^{7,15,16}

$$G(\mathbf{x}t, \mathbf{x}'t') = \sum_{ij} G_{ij}(t, t') \phi_i(\mathbf{x}) \phi_j^*(\mathbf{x}'). \quad (60)$$

When Eq. (60) is inserted in the expressions for the self-energy we obtain a basis set representation of the self-energy involving the matrices $G_{ij}(t, t')$ and the two-electron integrals which are given as integrals of orbital products with the two-body interaction v . All the quantities therefore become time-dependent matrices and all products are to be interpreted as matrix products. We will, however, suppress all matrix indices to display the temporal structure of the equations more clearly. Explicit expressions of the matrix form of the second Born and GW self-energy are given in Refs. 7, 15, and 16.

We start by discussing the time propagation of $G^>$ and $G^<$. Due to the symmetry relations [Eqs. (28) and (29)] we only need to calculate $G^>(t, t')$ for $t > t'$ and $G^<(t, t')$ for $t \leq t'$. From Eqs. (19) and (20) it then follows that $G^>$ must be time stepped in the first time argument and $G^<$ in the second one. We thus need to calculate $G^>(T+\Delta, t')$ and $G^<(t, T+\Delta)$ for a small time-step Δ , from the knowledge of $G^{\cong}(t, t')$ for $t, t' \leq T$. The symmetry relations (28) then immediately provide us with $G^>(t', T+\Delta)$ and $G^<(T+\Delta, t)$ as well. The time-stepping procedure is illustrated in Fig. 2 that displays the (t, t') -plane, in which at a given time T all the quantities inside the square with sides equal to T , are known. The time-step $G^<(t, T) \rightarrow G^<(t, T+\Delta)$ corresponds to a shift of the upper side of the time square with Δ , i.e., a shift from the solid to the dotted line in Fig. 2. Similarly the time-step

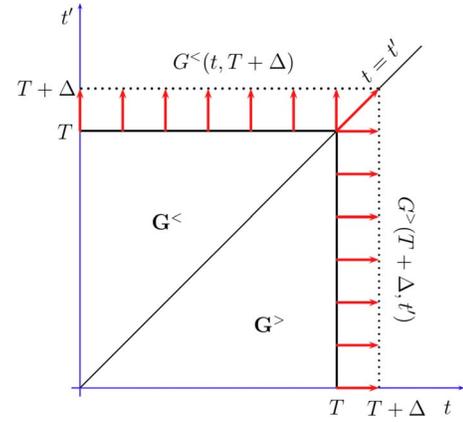


FIG. 2. (Color online) Time stepping in the (t, t') -plane. $G^>(t, t')$ is calculated for $t > t'$ and $G^<(t, t')$ is calculated for $t \leq t'$.

$G^>(T, t') \rightarrow G^>(T+\Delta, t')$ corresponds to a shift of the right-hand side of the time square with Δ . We further need to make a step $G^<(T, T) \rightarrow G^<(T+\Delta, T+\Delta)$ along the time diagonal $t = t'$. The propagation of $G^{\downarrow}(-i\tau, t)$ and $G^{\downarrow}(t, -i\tau)$ requires a time-step in the real time coordinate t for fixed imaginary time points τ .

Note that the right-hand sides of Eqs. (19)–(22) depend on the Green functions at the times $T+\Delta$, which are not known at time T . We therefore carry out the time-step $T \rightarrow T+\Delta$ twice. After taking the time-step for the first time, we recalculate the right-hand sides of Eqs. (19)–(22) and repeat the time-step $T \rightarrow T+\Delta$ using an average of the old and new collision and HF terms. Since the term $h^{\text{HF}}(t)$ in Eqs. (19)–(22) can attain large values, it is favorable to eliminate this term from the time-stepping equations. For each time-step $T \rightarrow T+\Delta$ we therefore absorb the term in a time-evolution operator of the form

$$U(t) = e^{-i\bar{h}^{\text{HF}}(T)t}, \quad (61)$$

where $\bar{h}^{\text{HF}}(T) = h(T+\Delta/2) + \Sigma^{\text{HF}}(T)$, where h is the one-body part of the Hamiltonian of Eq. (2). The one-body Hamiltonian $h(t)$ is explicitly known as a function of time and can be evaluated at half the time-step. The term Σ^{HF} is only known at time T and will be recalculated in the repeated time-step. In terms of the operator $U(t)$ of (61) we define new Green function matrices $g^x(x = \lessgtr, \uparrow\downarrow)$, as

$$G^{\lessgtr}(t_1, t_2) = U(t_1) g^{\lessgtr}(t_1, t_2) U^\dagger(t_2), \quad (62)$$

$$G^{\downarrow}(t_1, -i\tau_2) = U(t_1) g^{\downarrow}(t_1, -i\tau_2), \quad (63)$$

$$G^{\downarrow}(-i\tau_1, t_2) = g^{\downarrow}(-i\tau_1, t_2) U^\dagger(t_2). \quad (64)$$

We can now transform Eqs. (19)–(22) into equations for g^x . For instance, $g^>$ satisfies the equation

$$i\partial_t g^>(t, t') = U^\dagger(t) (h^{\text{HF}}(t) - \bar{h}^{\text{HF}}) G^>(t, t') U(t') + U^\dagger(t) I_1^>(t, t') U(t'). \quad (65)$$

Since $\bar{h}^{\text{HF}} \approx h^{\text{HF}}(t)$ for times $T \leq t \leq T+\Delta$, we can neglect for these times the first term on the right-hand side of Eq. (65). We then find

$$\begin{aligned}
G^>(T+\Delta, t_2) &= U(T+\Delta)g^>(T+\Delta, t_2)U^\dagger(t_2) \\
&= U(T+\Delta) \left[g^>(T, t_2) + \int_T^{T+\Delta} dt \partial_t g^>(t, t_2) \right] U^\dagger(t_2) \\
&\approx U(\Delta)G^>(T, t_2) - iU(\Delta) \left\{ \int_T^{T+\Delta} dt e^{i\bar{h}^{\text{HF}}(t-T)} \right\} I_1^>(T, t_2) = U(\Delta)G^>(T, t_2) - V(\Delta)I_1^>(T, t_2), \quad (66)
\end{aligned}$$

where $V(\Delta)$ is defined as

$$V(\Delta) = (\bar{h}^{\text{HF}})^{-1} [1 - e^{-i\bar{h}^{\text{HF}}\Delta}]. \quad (67)$$

Similarly for $G^<$, which is propagated using Eq. (20), we find the equation

$$G^<(t_1, T+\Delta) = G^<(t_1, T)U^\dagger(\Delta) - I_2^<(t_1, T)V^\dagger(\Delta). \quad (68)$$

For time stepping along the time diagonal we use

$$i\partial_t G^<(t, t) = [h^{\text{HF}}(t), G^<(t, t)] + I_1^<(t, t) - I_2^<(t, t), \quad (69)$$

which follows directly from a combination of the equations for $G^<$ of Eqs. (19) and (20). The corresponding equation for $g^<(t, t)$ on the time diagonal then becomes

$$\begin{aligned}
i\partial_t g^<(t, t) &= U^\dagger(t)[h^{\text{HF}}(t) - \bar{h}^{\text{HF}}, G^<(t, t)]U(t) \\
&\quad + U^\dagger(t)(I_1^<(t, t) - I_2^<(t, t))U(t). \quad (70)
\end{aligned}$$

From this equation we then obtain

$$\begin{aligned}
G^<(T+\Delta, T+\Delta) &= U(T+\Delta)g^<(T+\Delta, T+\Delta)U^\dagger(T+\Delta) \\
&= U(\Delta)G^<(T, T)U^\dagger(\Delta) - iU(\Delta) \\
&\quad \times \left[\int_0^\Delta dt U^\dagger(t)I_{12}U(t) \right] U^\dagger(\Delta), \quad (71)
\end{aligned}$$

where we defined $I_{12} = I_1^<(T, T) - I_2^<(T, T)$. By using the operator expansion

$$e^A B e^{-A} = B + [A, B] + \frac{1}{2}[A, [A, B]] + \frac{1}{3}[A, \frac{1}{2}[A, [A, B]]] + \dots, \quad (72)$$

it follows that

$$-i \int_0^\Delta dt U^\dagger(t)I_{12}U(t) = \sum_{n=0}^{\infty} C^{(n)}, \quad (73)$$

where

$$C^{(n)} = \frac{i\Delta}{n+1} [\bar{h}^{\text{HF}}, C^{(n-1)}], \quad (74)$$

and $C^{(0)} = -i\Delta I_{12}$. If we insert Eq. (73) into Eq. (71) we finally obtain

$$G^<(T+\Delta, T+\Delta) = U(\Delta) \left[G^<(T, T) + \sum_{n=0}^{\infty} C^{(n)} \right] U^\dagger(\Delta). \quad (75)$$

We found that keeping terms for $n \leq 3$ only, yields sufficient accuracy. We now consider the time propagation for the

mixed real and imaginary time Green functions. For g^l we have the equation

$$\begin{aligned}
i\partial_t g^l(t, -i\tau) &= U(t)^\dagger(h^{\text{HF}}(t) - \bar{h}^{\text{HF}})G^l(t, -i\tau) \\
&\quad + U(t)^\dagger l^l(t, -i\tau). \quad (76)
\end{aligned}$$

This yields, similarly as in Eqs. (66) and (68)

$$G^l(T+\Delta, -i\tau_2) = U(\Delta)G^l(T, -i\tau_2) - V(\Delta)l^l(T, -i\tau_2). \quad (77)$$

Finally, for G^l we have

$$G^l(-i\tau_1, T+\Delta) = G^l(-i\tau_1, T)U(\Delta)^\dagger - l^l(-i\tau_1, T)V(\Delta)^\dagger. \quad (78)$$

Equations (66), (68), (71), (77), and (78) form the basis of the time-stepping algorithm. At each time-step, it requires the construction of the step operators $U(\Delta)$ and $V(\Delta)$ and therefore the diagonalization of \bar{h}^{HF} for every time-step. As mentioned before, the right-hand sides of Eqs. (19)–(22) depend on the Green functions at the times $T+\Delta$ which are not known at time T . We therefore carry out the time-step $T \rightarrow T+\Delta$ twice. The procedure is as follows:

- (1) The collision integrals and \bar{h}^{HF} at time T are calculated from the Green functions for times $t, t' \leq T$.
- (2) A step in the Green function $G(T) \rightarrow G(T+\Delta)$ is taken according to Eqs. (66), (68), (71), (77), and (78).
- (3) New collision integrals $I_1^>(T+\Delta, t), I_2^>(t, T+\Delta), l^l(T+\Delta, -i\tau)$ and $l^l(-i\tau, T+\Delta)$ are calculated by inserting the new Green functions for times $t, t' \leq T+\Delta$ into Eqs. (24)–(27).
- (4) The values of the collision integrals and the HF self-energy are approximated by $\bar{I} = (I(T) + I(T+\Delta))/2$ and $\bar{\Sigma}^{\text{HF}} = (\Sigma^{\text{HF}}(T) + \Sigma^{\text{HF}}(T+\Delta))/2$ where $I(T)$ and $I(T+\Delta)$ are the collision terms calculated under points (1) and (3).
- (5) The Green function is then propagated from $G(T) \rightarrow G(T+\Delta)$ using the average values \bar{I} and $\bar{h}^{\text{HF}} = h(T+\Delta/2) + \bar{\Sigma}^{\text{HF}}$ in Eqs. (66), (68), (71), (77), and (78).

This concludes the general time-stepping procedure for the Green functions.

We finally consider the calculation of $\tilde{W}^<$ and \tilde{W}^l from Eqs. (52) and (53). As a consequence of the symmetry relation (50), we only need to calculate $\tilde{W}^<(t, t')$ for $t < t'$. In a time-step from T to $T+\Delta$ we need to calculate $\tilde{W}^<(t, T+\Delta)$

for $t \leq T + \Delta$ from the known values of $\tilde{W}^<(t, T)$ for $t \leq T$. The first term on the right-hand side of Eq. (52) can be calculated directly from $G^<(t, T + \Delta)$ and $G^>(T + \Delta, t)$. However, the last term $X^<(t, T + \Delta)$ of Eq. (52) depends on the, still undetermined, values $\tilde{W}^<(t, T + \Delta)$. We therefore employ an iterative scheme. As a first guess for $\tilde{W}^<(t, T + \Delta)$ we take $\tilde{W}^<(t, T + \Delta) = \tilde{W}^<(t, T)$ for $t \leq T$ and $\tilde{W}^<(T + \Delta, T + \Delta) = \tilde{W}^<(T, T)$. We therefore use the values of $\tilde{W}^<$ on the known sides of the time square at time T (solid lines in Fig. 2) as initial guesses for the stepped sides (dotted lines in Fig. 2) at $T + \Delta$. As an initial guess for the value of $\tilde{W}^<$ at the new diagonal point $(T + \Delta, T + \Delta)$, we take the value at the previous diagonal point (T, T) . We then calculate the quantity $X^<(t, T + \Delta)$ for $t \leq T + \Delta$ and obtain a new value for $\tilde{W}^<(t, T + \Delta)$ from Eq. (52). This value is then inserted back into the right-hand side of Eq. (52) and the process is repeated until convergence is reached. Similarly we initialize $\tilde{W}^>(T + \Delta, -i\tau) = \tilde{W}^>(T, -i\tau)$ and solve Eq. (53) in the same manner as for $\tilde{W}^<$.

This concludes our derivation of the time-stepping algorithm of the KB equations. The propagation method described here has been used in two recent letters^{7,11} where also values for the numerical parameters are given. It is clear that the choice of these parameters depends strongly on the type of system considered, and on the strength of the applied external fields.

V. SUMMARY AND CONCLUSIONS

We presented a detailed account of the KB equations and discussed in detail their structure, initial conditions, and symmetries. We developed an algorithm for the time propagation of the KB equations in which the symmetry relations for the Green functions were used to reduce the set equations that needed to be solved. In two recent letters^{7,11} we applied the method to the case of atoms and molecules in external time-dependent fields and to the case of transient transport dynamics of double quantum dots. We therefore conclude that time propagation of the KB equations can be used as a practical method to calculate the nonequilibrium properties of a wide variety of many-body quantum systems, ranging from atoms and molecules to quantum dots and quantum wells. Moreover, the present work can be readily extended to other Green function formalisms, such as the Nambu

formalism^{28,29} for superconducting systems. Also future extension to bosonic systems is straightforward. Work along these lines is in progress.

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