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Steady state conductance in a double quantum dot array:
The nonequilibrium equation-of-motion Green function approach

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We study steady state transport through a double quantum dot array using the equation-of-motion approach to the nonequilibrium Green functions formalism. This popular technique relies on uncontrolled approximations to obtain a closure for a hierarchy of equations; however, its accuracy is questioned. We focus on 4 different closures, 2 of which were previously proposed in the context of the single quantum dot system (Anderson impurity model) and were extended to the double quantum dot array, and develop 2 new closures. Results for the differential conductance are compared to those attained by a master equation approach known to be accurate for weak system-leads couplings and high temperatures. While all 4 closures provide an accurate description of the Coulomb blockade and other transport properties in the single quantum dot case, they differ in the case of the double quantum dot array, where only one of the developed closures provides satisfactory results. This is rationalized by comparing the poles of the Green functions to the exact many-particle energy differences for the isolate system. Our analysis provides means to extend the equation-of-motion technique to more elaborate models of large bridge systems with strong electronic interactions.

I. INTRODUCTION

The interest in transport through conjugated molecules has grown in recent years in light of their potential applications in electronic and optoelectronic devices.1, 2 While certain transport properties can be treated within a noninteracting picture via the tight binding approximation combined with the Landauer formalism,3 often the description of transport requires the inclusion of many-body electron-electron and/or electron-phonon correlations.4 For very simple and small systems, introducing such correlations can be done, for example, by means of time-dependent numerical renormalization-group techniques,5-6 many-body wavefunction approaches,7 diagrammatic techniques to real time path integral formulation,8-11 or reduced dynamic methods.12, 13 The treatment of correlations becomes a greater theoretical challenge in systems with many electronic states driven away from equilibrium,14, 15 where the computational cost of numerical techniques increases rapidly beyond current capabilities.

A central framework dealing with transport in large systems is the nonequilibrium Green functions (NEGF) formalism.16-19, 41-42 The equation-of-motion (EOM) method is one of the more basic ways to calculate the Green functions (GF) of an interacting quantum system. Its main advantages are the simplicity and relatively mild scaling with the number of electrons, under simple truncation schemes.20 The EOM nonequilibrium Green function formalism provides a qualitative description of transport phenomena in strongly correlated systems, such as the Coulomb blockade effect21-23 and the Kondo effect in quantum dots.24, 25 However, questions regarding the validity of the EOM approach have been raised.26, 27 For example, it has been shown to violate the Friedel sum rule28 near the Kondo regime26 and basic Green function symmetry relations away from the Kondo regime.27 In the latter case, symmetry relations can be restored and at least for the Anderson impurity model,29 the approach recovers the Kondo peaks and provides a quantitative description of resonant transport.27

In this paper, we study the role of different approximate closures to the EOM of the NEGF formalism on steady state properties (namely, the differential conductance) for a double quantum dot (QD) array, coupled to two macroscopic leads (an extended Hubbard model,30, 31 also known as the double Anderson model32). Although general, this model can be used to study transport through single diatomic molecules (where each atom is represented by its conducting orbital)33 and even larger molecules34, 35 or structures.40 Four closures are examined; two already proposed23, 33 (approximations 1 and 4 described in Secs. II B 1 and II B 4, respectively) and two developed in this work (approximations 2 and 3 described in Secs. II B 2 and II B 3, respectively). The results obtained from the different closures were compared to the results attained using a many-particle Master Equation (ME) approach34 adequate for weak hybridization (system-leads couplings) and high temperatures.35, 36 In contrast to the simplest case of a single site model (Anderson impurity model) in which different closures beyond the simplest Hartree approximation scheme37 yield very similar transport results,20 (steady state current and differential conductance as a function of the applied bias voltage) at high temperatures, we show that this is not the case for the double QD array, where different closures yield very
different steady state currents and differential conductance curves.

The performance of the different closures is analysed in terms of the poles of the GFs in comparison to the exact many-body result for the isolated system. We find that one of the closures developed in this work provides the most accurate description of the poles and also the best overall agreement with the ME approach for all parameters studied in this work. While these results are encouraging, a word of caution is in place. It is clear that the conclusions drawn from the performance of the different closures for the single site model cannot be extended directly to the two site model, in analogy, a suitable closure for the two site model may fail in the larger systems. Thus, the study of larger arrays of QDs will require analysis along the lines sketched in this work.

The paper is organized as follows: in Sec. II, we present the double QD model Hamiltonian, provide a short description of the equation-of-motion technique and a detailed description of the different approximate closures to the EOM. Sequential labeling of the different approximate closures refers to the order of the closure. Finally, a brief summarization of the ME approach is provided as well. Results and discussion are given in Sec. III for the cases of the symmetric and asymmetric bridges. In Sec. IV we conclude.

II. THEORY

A. Model Hamiltonian

We consider a system of two coupled QDs array connected to two macroscopic leads, as sketched in Figure 1. The Hamiltonian has the following general form:

$$\hat{H} = \hat{H}_B + \hat{H}_S + \hat{H}_i,$$

(1)

with $\hat{H}_B$ describing the macroscopic leads (left and right contacts), $\hat{H}_S$ describes the system of interest, and $\hat{H}_i$ is the interaction Hamiltonian between the system and the leads. The leads (left ($L$) and right ($R$)) are modeled as infinite non-interacting fermionic baths, and are assumed to be each at its own equilibrium, characterized by chemical potentials $\mu_L$ and $\mu_R$, where the difference $\mu_L - \mu_R = e\Phi$ is the applied voltage bias. The leads’ Hamiltonian is given by

$$\hat{H}_B = \sum_{\sigma,k \in \{L,R\}} \epsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma},$$

(2)

where $\epsilon_{k\sigma}$ is the energy of a free electron in the left or right lead, in momentum state $k$ and spin $\sigma$. The operators $c_{k\sigma}$/$c_{k\sigma}^\dagger$ are the annihilation/creation operators of such an electron. The double QD system is described by an extended Hubbard model

$$\hat{H}_S = \sum_{\sigma,m \in \{\alpha,\beta\}} \epsilon_{m\sigma} n_{m\sigma} + U \sum_m n_{m\uparrow} n_{m\downarrow} + V \sum_{\sigma,\sigma',\beta} n_{\alpha\sigma} n_{\beta\sigma'} + h \sum_{\sigma} (d_{\alpha\sigma}^\dagger d_{\beta\sigma} + h.c.),$$

(3)

where $n_{\alpha\sigma} = d_{\alpha\sigma}^\dagger d_{\alpha\sigma}$ is the number operator of the electron occupying site (dot) $\alpha$ with spin $\sigma$ and energy $\epsilon_{\alpha\sigma}$, $U$ is the repulsion energy between two electrons on the same site with opposite spins (intra-dot repulsion), $V$ is the repulsion energy between two electrons on different sites (inter-dot repulsion), and $h$ is the coupling strength for electron hopping between the two sites. The interaction between the system and the contacts is simply given by the tunnelling Hamiltonian

$$\hat{H}_i = \sum_{\sigma,k \in L} (t_{\sigma\alpha}^c c_{k\sigma}^\dagger d_{\alpha\sigma} + h.c.) + \sum_{\sigma,k \in R} (t_{\sigma\beta}^c c_{k\sigma}^\dagger d_{\beta\sigma} + h.c.).$$

(4)

The parameter $t_{\sigma\alpha}^c$ represents the coupling strength (hybridization) between the system and the leads, and the index $m$ runs over the site index $\{\alpha, \beta\}$.

B. Equation of motion

The above model consists of an interacting system coupled to two electron reservoirs with specified chemical potentials and temperatures. In order to obtain a solution to this many-body out of equilibrium problem, we resort to the EOM approach within the NEGF formalism. We begin by defining the contour ordered GF, $\hat{G}_{\sigma\beta}^c(t_2,t_1) = -\frac{i}{\hbar} (T_c \Psi_{\sigma}(t_2) \hat{\Psi}_{\beta}^\dagger(t_1))$, where $\Psi/\Psi^\dagger$ are the system’s annihilation.creation field operators, and $T_c$ is the contour time ordering operator. The EOM for the contour ordered GF is obtained from the Heisenberg EOM for a Heisenberg operator $\hat{A}(t) = i\hbar \{\hat{H}(t), \hat{A}(t)\} + \frac{\partial}{\partial t} \hat{A}(t)$, where $[\hat{A}, \hat{B}] = \hat{A}\hat{B} - \hat{B}\hat{A}$. A full description of the system requires the knowledge of the retarded, advanced, and lesser (distribution) GFs

$$\hat{G}_{\sigma\beta}^r(t_2,t_1) = -\frac{i}{\hbar} \theta(t_2 - t_1) \langle \hat{\Psi}_{\sigma}(t_2), \hat{\Psi}_{\beta}^\dagger(t_1) \rangle,$$

$$\hat{G}_{\sigma\beta}^a(t_2,t_1) = \frac{i}{\hbar} \theta(t_1 - t_2) \langle \hat{\Psi}_{\sigma}(t_1), \hat{\Psi}_{\beta}^\dagger(t_2) \rangle,$$

(5)

$$\hat{G}_{\sigma\beta}^<^>(t_2,t_1) = \frac{i}{\hbar} \langle \hat{\Psi}_{\sigma}^r(t_1), \hat{\Psi}_{\beta}^a(t_2) \rangle,$$

where $[A, B]$ is the anti-commutator of $A$ and $B$. These real time GFs can be extracted from the contour ordered GF using

\[\text{FIG. 1. A sketch of the double QD bridge. See main text for the definition of all quantities.}\]
the Langreth rules. Except for very simple models (e.g., see Refs. 54 and 55), the EOMs of the NEGF will produce “new” and higher order GFs that need to be evaluated. In general this leads (after a few iterations) to a non-tractable hierarchy of equations. To obtain a working closure one has to truncate the resulting set of equations and/or decouple the higher order GFs and express them via lower order ones. Recently, it has been shown that such a procedure may lead to symmetries violations that the GFs must obey by definition27 and a routine to restore back the symmetries was suggested.27 In what follows we examine 4 different closures to the EOMs of the NEGF for the Hamiltonian discussed in Sec. II A, and apply the symmetry restoration scheme to circumvent the inherent flaw of the EOMs approach. Our starting point is the contour ordered GF,
\[
\hat{G}_{\alpha\beta}(t, t') = -\frac{i}{\hbar}(T_{d\alpha\sigma}(t) d_{\beta\sigma}^\dagger(t')).
\]  
In steady state, the NEGF depends only on the time difference \( \tau = t - t' \), and thus would have a simpler representation in Fourier space. The resulting EOM for the single particle GF is given by
\[
G_{\alpha\beta}(\omega) = \left( \hbar \omega + \varepsilon_{\beta\sigma} - V - \Sigma_{\alpha\sigma}^{0}(\omega) \right)^{-1} \times \left( \delta_{\alpha\beta} + h\Sigma_{\alpha\beta}(\omega) \right) + 1 \times \left( \delta_{\alpha\beta} + h\Sigma_{\alpha\beta}(\omega) \right) = -\frac{1}{\hbar}(T_{d\alpha\sigma}(t) d_{\beta\sigma}^\dagger(t)) \frac{(\omega - \varepsilon_{\beta\sigma})}{(\omega - \varepsilon_{\alpha\sigma})},
\] and
\[
\Sigma_{\alpha\beta}^{\sigma}(\omega) = \left( \hbar \omega - \varepsilon_{\beta\sigma} - V - \Sigma_{\alpha\sigma}^{\delta}(\omega) \right)^{-1} \times (\delta_{\alpha\beta} + h\Sigma_{\alpha\beta}(\omega) - h\Sigma_{\alpha\beta}^{\delta}(\omega)) + 1 \times (\delta_{\alpha\beta} + h\Sigma_{\alpha\beta}^{\delta}(\omega) + U\Sigma_{\alpha\beta}^{\delta}(\omega) + \Sigma_{\alpha\beta}^{\delta}(\omega) + \sum_{k} \left( \frac{\delta_{\alpha\beta}}{T_{d\alpha\sigma}}(\omega) - \frac{\delta_{\alpha\beta}}{T_{d\alpha\sigma}}(\omega) \right) + \frac{1}{T_{d\alpha\sigma}}(\omega)),
\] In the above equations, \( \Sigma_{\alpha\beta}^{\sigma}(\omega) \), \( \Sigma_{\alpha\beta}^{\delta}(\omega) \), \( \Sigma_{\alpha\beta}^{\tag{11}}(\omega) \), and \( \Sigma_{\alpha\beta}^{\tag{12}}(\omega) \) are the Fourier transforms of \( \Sigma_{\alpha\beta}^{\tag{11}}(t, t') \), \( \Sigma_{\alpha\beta}^{\tag{12}}(t, t') \), \( \Sigma_{\alpha\beta}^{\tag{11}}(t, t') \), and \( \Sigma_{\alpha\beta}^{\tag{12}}(t, t') \), respectively. To continue, one has to formulate the equations for these new GFs, which in turn will lead to other (higher order) GFs. This infinite hierarchy of equations needs to be truncated at a certain level, a process which is referred to as “closure.” In general, closures cannot be improved systematically. Furthermore, it is often difficult to assess, a priori, the accuracy of a given closure. We now discuss several different closures which are physically motivated, tractable, and some are commonly used in the context of transport.

1. Approximation-1

Following the derivation given in Ref. 23, the following approximations are made: (a) all 3-particle GFs are set to zero, (b) simultaneous tunnelling of electrons of opposite spins are neglected, (c) GFs mixing leads and system operators are decoupled so \( \hat{G}_{\alpha\beta}(t, t') = \frac{1}{\hbar}(T_{d\alpha\sigma}(t) d_{\beta\sigma}^\dagger(t')) \approx \frac{1}{\hbar} \int dt_{r} \hat{G}_{\alpha\beta}(t, t_{r}) T_{d\alpha\sigma}(t_{r}) d_{\beta\sigma}^\dagger(t_{r}) \), where \( \frac{1}{\hbar} \) is constant (as is the case in steady state). (d) The remaining 2-particle GFs of the form \( \hat{G}_{\alpha\beta}(t, t') \) are decoupled \( \hat{G}_{\alpha\beta}(t, t') = (n_{\beta\sigma}(t)) \hat{G}_{\alpha\beta}(t, t') \). Assumption (c) is equivalent to treating the coupling to the leads up to the second order with respect to \( t_{r}^{\alpha\beta} \). It neglects processes necessary to qualitatively capture the Kondo effect yet results are predicted to be reliable for temperatures above the Kondo temperature \( (T_{K}) \). The resulting equations are given by (for brevity, we omit the implicit dependence on \( \omega \)),
\[
G_{\alpha\alpha}(\omega) = \left( \hbar \omega - \varepsilon_{\alpha\alpha} - U - \Sigma_{\alpha\alpha}^{0}(\omega) \right)^{-1} \times (1 + hG_{\beta\alpha}^{\sigma} + UG_{\alpha\alpha}^{\sigma} + VG_{\beta\alpha}^{\sigma} + VG_{\beta\alpha}^{\sigma}),
\] and
\[
G_{\beta\alpha}(\omega) = \left( \hbar \omega - \varepsilon_{\beta\alpha} - U - \Sigma_{\beta\alpha}^{0}(\omega) \right)^{-1} \times (hG_{\beta\alpha}^{\sigma} + UG_{\beta\alpha}^{\sigma} + VG_{\beta\alpha}^{\sigma} + VG_{\beta\alpha}^{\sigma}),
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - U - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \hbar \langle n_{\beta \sigma} \rangle G_{\alpha \alpha}^{\sigma \sigma},
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - V - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \hbar \langle n_{\alpha \sigma} \rangle G_{\alpha \alpha}^{\sigma \sigma},
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - V - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \langle \alpha_{\beta \sigma} \rangle - \langle d_{\alpha \sigma}^1 d_{\beta \sigma} \rangle,
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - U - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \langle n_{\alpha \sigma} \rangle G_{\alpha \alpha}^{\sigma \sigma},
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - V - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \langle n_{\beta \sigma} \rangle + \langle n_{\beta \sigma} \rangle G_{\beta \beta}^{\sigma \sigma}.
\]

In general the GFs depend on the expectation values of \(\langle n_{\gamma \tau} \rangle = \langle d_{\gamma \tau}^1 d_{\gamma \tau} \rangle\) and \(\langle d_{\alpha \sigma}^1 d_{\beta \tau} \rangle\), which are determined by means of the lesser GF,

\[
\langle d_{\alpha \sigma}^1 d_{\beta \tau} \rangle = -\frac{i \hbar}{2\pi} \int_{-\infty}^{\infty} (G_{\alpha \beta}^{\sigma \sigma}(\omega))^{-1} d\omega,
\]

thus, a self-consistent calculation is required.

2. Approximation-3

A seemingly better approximation scheme is one that relaxes the last mean-field approximation (assumption “d”) described in Sec. II B 1 and the 2-particle GFs of the form 
\[
G_{\alpha \beta}^{\sigma \sigma}(t, t') = -\frac{i}{\hbar} (T_{C_{\alpha \beta}}(t) d_{\beta \sigma}(t) d_{\gamma \tau}^1(t'))
\]
are not decoupled but treated fully. Thus, while Eq. (11) remains the same, the equations for the 2-particle GFs will now be given by

\[
(\hbar \omega - \epsilon_{\beta \sigma} - V - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \hbar \epsilon_{\beta \sigma}^{\sigma \sigma},
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - V - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \hbar \epsilon_{\alpha \sigma}^{\sigma \sigma} - \langle d_{\alpha \sigma}^1 d_{\beta \sigma} \rangle,
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - U - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \hbar \epsilon_{\alpha \sigma}^{\sigma \sigma},
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - V - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \langle n_{\beta \sigma} \rangle + \hbar \epsilon_{\alpha \sigma}^{\sigma \sigma},
\]
\[
(\hbar \omega - \epsilon_{\beta \sigma} - V - \Sigma_{\beta \sigma}^0) G_{\alpha \beta}^{\sigma \sigma} = \langle n_{\beta \sigma} \rangle + \hbar \epsilon_{\beta \sigma}^{\sigma \sigma}.
\]

As noted above, the retarded, advanced, and lesser GFs can now be evaluated using Langreth theorem,\(^{53}\) and the expectation values \(\langle n_{\gamma \tau} \rangle = \langle d_{\gamma \tau}^1 d_{\gamma \tau} \rangle\) and \(\langle d_{\alpha \sigma}^1 d_{\beta \tau} \rangle\) are determined via the lesser GF.

3. Approximation-3

A more complete treatment of the 2nd order GFs requires relaxing assumption “b” in addition to assumption “d”, described in Sec. II B 1. Namely, we attend to the 2-particle GFs that describe simultaneous tunnelling of electrons of opposite spins in the double QD system, 
\[
G_{\alpha \beta}^{\sigma \sigma}(t, t') = \frac{1}{\hbar} (T_{C_{\alpha \beta}}(t) d_{\beta \sigma}(t) d_{\gamma \tau}^1(t')).
\]
Again, the only changes are in the equations for the 2-particle GFs, and the resulting equations are given by

\[
G_{\alpha \beta}^{\sigma \sigma} = \frac{1}{\hbar} \epsilon_{\alpha \sigma}^{\sigma \sigma} + \langle d_{\alpha \sigma}^1 d_{\beta \sigma} \rangle,
\]
\[
G_{\alpha \beta}^{\sigma \sigma} = \langle n_{\beta \sigma} \rangle + \hbar \epsilon_{\alpha \sigma}^{\sigma \sigma},
\]
\[
G_{\alpha \beta}^{\sigma \sigma} = \langle n_{\beta \sigma} \rangle + \hbar \epsilon_{\beta \sigma}^{\sigma \sigma}.
\]

This case is not different from the previous two in the sense that a self-consistent treatment is required.

4. Approximation-4

Finally, we follow the derivation of Ref. 33. Here, the following approximations are made: (a) simultaneous tunnelling of electrons of opposite spins are neglected, (b) GFs mixing leads and system operators are decoupled so 
\[
\approx -\frac{1}{\hbar} \int dt \langle d_{\gamma \tau}^1(t) \rangle \langle n_{\beta \sigma} \rangle (\hbar)_{\beta \gamma}^{\gamma \sigma},
\]
\[
\approx -\frac{1}{\hbar} \int dt \langle d_{\gamma \tau}^1(t) \rangle \langle d_{\beta \sigma}^1(t) \rangle \langle n_{\beta \sigma} \rangle (\hbar)_{\beta \gamma}^{\gamma \sigma},
\]
\[
\approx -\frac{1}{\hbar} \int dt \langle d_{\gamma \tau}^1(t) \rangle \langle d_{\beta \sigma}^1(t) \rangle \langle n_{\beta \sigma} \rangle (\hbar)_{\beta \gamma}^{\gamma \sigma},
\]
\[
\approx -\frac{1}{\hbar} \int dt \langle d_{\gamma \tau}^1(t) \rangle \langle d_{\beta \sigma}^1(t) \rangle \langle n_{\beta \sigma} \rangle (\hbar)_{\beta \gamma}^{\gamma \sigma}.\]

The resulting equations are given
C. Master equations

The results obtained from the different closures were compared to the results attained using a many-particle ME approach. For completeness, we briefly outline the ME formalism and provide expressions for the transition rates for the double QD model.

In the many-particle picture, the system has different probabilities $p_i$ of being in one of its $2^N$ possible eigenstates ($N$ being the maximum occupancy of the system, e.g., in our case of the double QD system $N = 4$, and there are 16 many-particle states). Normalization requires that the probabilities sum up to one

$$\sum_{i=1}^{2^N} p_i = 1,$$

and in steady state there is no net current into or out of any state, i.e.,

$$\sum_{j=1}^{2^N} R_{j \to i} \cdot p_i = \sum_{j=1}^{2^N} R_{i \to j} \cdot p_j,$$

where $R_{j \to i}$ is the rate of transition from the many-particle state $|n_1, i\rangle$ to $|n_2, j\rangle$ (the $j$th out of the $n_2$-particle states).

The many-particle states are incurred by diagonalizing the system’s Hamiltonian $H_L$, while the rate constants are calculated assuming a specific model for the interactions between our system and its environment (the leads). Knowing the transition rates the probabilities can be evaluated and the steady state current is then given by

$$I_L = -e \sum_{ij} (n_i - n_j) R_{i \to j} \cdot p_j,$$

where $n_i$ is the occupation number of state $|n, i\rangle$ and $R_{i \to j}$ is the rate constant that represents the part of the total transition rate associated with the left lead. For sequential tunneling we assume transitions only between many-particle states that obey $|n_i - n_j| = 1$. In the weak coupling regime the interaction between the system and the leads is treated perturbatively and the rates are given by the Fermi golden rule

$$R_{i \to j}^L = \sum_{\sigma, \epsilon_k} \frac{2\pi |r^\sigma_{k\epsilon}|^2 \delta (\epsilon - \epsilon_{k\sigma})}{\hbar}$$

$$\times (f_L(E_{ij} - \mu_L) |n_2, j| d^\sigma_{a\epsilon} |n_1, i\rangle)^2$$

$$+ (1 - f_L(E_{ji} - \mu_L)) |n_1, i| d^\sigma_{a\epsilon} |n_2, j\rangle|^2).$$

In the above, $E_{ij} = E_i - E_j$, is the difference in energy between the many-particle states $|n_1, i\rangle$ and $|n_2, j\rangle$, and $f_\epsilon(\epsilon - \mu_\epsilon)$ is the Fermi–Dirac distribution. Defining the matrix coupling of the system to the left reservoir, $\Gamma_L$, with elements $(\Gamma_{\alpha\beta}\sigma)_{k\epsilon} = \frac{2\pi}{\hbar} |\sum_{k\epsilon} \delta (\epsilon - \epsilon_{k\sigma})| e^{\sigma}_{k\epsilon} |^2$, we can rewrite equation (20) as

$$R_{i \to j}^L = \sum_{\sigma, \epsilon_k} \frac{(\Gamma_{\alpha\beta}\sigma)_{k\epsilon}}{\hbar} \times (f_L(E_{ij} - \mu_L) |n_2, j| d^\sigma_{a\epsilon} |n_1, i\rangle)^2$$

$$+ (1 - f_L(E_{ji} - \mu_L)) |n_1, i| d^\sigma_{a\epsilon} |n_2, j\rangle|^2).$$

III. RESULTS AND DISCUSSION

At this point we wish to examine the different approximations and find which one leads to a system GF that describes the double QD more accurately or at least qualitatively. As we are interested in transport properties of a system weakly coupled to the macroscopic leads we will compare the results obtained from the different approximations to the ones calculated using the many-particle ME approach. Under these assumptions, the ME is believed to be fairly accurate. We wish to note that the EOM approach is not limited to the weak coupling case. As a measure of the quality of the approximations we chose to calculate the differential conductance, $dI/d\Phi$. In the many-particle picture, in the wide band limit (where the interaction with the leads only broadens the energy levels of the system without introducing any spectral shift), we expect to observe peaks in the differential conductance at values of the bias voltage that correspond to $\mu_L + \epsilon_L = \pm e\Phi/2 \approx \Delta E(N) = E(N) - E(N - 1)$, where $E_L$ is the equilibrium Fermi energy of the electrodes (throughout taken to be zero), $E(N)$ is the energy of the many-particle state with $N$ electrons of the unperturbed system, and $\Phi$ is the applied voltage. The voltage can be applied symmetrically to both leads (i.e., $\mu_L = E_f + e\Phi/2$ and $\mu_R = E_f - e\Phi/2$), or asymmetrically ($\mu_L = E_f + e\Phi$ and $\mu_R = E_f$). In the present study we have used the symmetric version.
The differential conductance is derived from differentiating the steady state current with respect to the bias voltage and was evaluated from the Meir-Wingreen formula\(^{61}\)

\[
I = \frac{ie}{2\pi\hbar} \int d\omega \text{Tr}(f_L(\epsilon - \mu_L) G_L(\epsilon) \\
\times (G'(\epsilon) G^{-\dagger}(\epsilon)) + \text{Tr}[\Gamma_L G^{-\dagger}(\epsilon)]) \tag{22}
\]

In the above, \(G'(\epsilon)\), \(G^\dagger(\epsilon)\), and \(G^{-\dagger}(\epsilon)\) are the retarded, advanced, and lesser GFs, respectively. The resulting EOMs were solved self-consistently in Fourier space with a frequency discretization of \(d\omega = 0.0005U\) over 32 768 grid points. Depending on the approximation, 15–150 self-consistent iterations were required to converge the results. Convergence was declared when the population values \(\langle n_{\text{mf}} \rangle\) at subsequent iterations did not change within a predefined tolerance value chosen to be \(10^{-6}\). For each set of calculations symmetrization routine was applied to restore the symmetry relations of the GFs.\(^{27}\)

A. Symmetric bridge

The transport through the double QD system can be classified into symmetric and asymmetric bridge setups, with or without inter-dot repulsion term, \(V\). In this section, we first consider the symmetric setup in which \(\epsilon_{\alpha\uparrow} = \epsilon_{\alpha\downarrow} = \epsilon_{\beta\uparrow} = \epsilon_{\beta\downarrow} = \epsilon = 0.35U\). The remaining model parameters were taken to be \(\Gamma^\dagger_{La} = \Gamma^\dagger_{La} = \Gamma^\dagger_{Rh} = \Gamma^\dagger_{Rb} = 0.015U\), \(\Gamma^\dagger_{Ld} = \Gamma^\dagger_{Ld} = \Gamma^\dagger_{Rd} = \Gamma^\dagger_{Rd} = 0\), and \(\beta^{-1} = U/40\). To make the connection with chemical systems, one can consider the on-site repulsion \(U\) to be of the order of 1–4 eV.\(^{30}\) This implies that \(\Gamma\) is of the order of 0.015–0.06 eV. In the case of the symmetric bridge the system’s single particle energy levels are around 0.35–1.4 eV above the leads’ Fermi energy and the inter-site repulsion (exciton binding energy) \(V\) is of the order of 0.8–3.2 eV.

In Figure 2, we plot the differential conductance for a symmetric bridge for different values of the hopping term \(h\). We set the inter-dot repulsion \(V = 0\). The black curves (solid line) are the results obtained by fully diagonalizing the bare system \(\hat{H}\) (and solving the ME). The other curves represent the outcome of the NEGF formalism within the different closure approximations. We also label the different peaks in the differential conductance, obtained via the ME approach, with the corresponding transitions between many-body states, i.e., \(|0\rangle \rightarrow |1\rangle\) corresponds to transitions from an empty system to a system with a single electron, etc. In the single particle GF formalism, peaks in the differential conductance will occur at the poles of the calculated GF. Hence, a good approximation is one that will produce single particle GF with poles at the position of the many-particle transitions.

For the smallest value of \(h\), we find that all approximations agree qualitatively with the ME approach. When the value of \(h\) is increased it is clear that approximation 1 (red circles) breaks down, implying that this simple closure is insufficient to describe strong hopping between the quantum dots. Approximations 2 and 3 (green diamonds and blue triangles, respectively) do agree with the ME, but “miss” certain conductance peaks (e.g., the peaks at \(\Phi_U \approx 0.75\) in the upper right panel, \(\Phi_U \approx 0.5\) in the lower left panel, and the peak at \(\Phi_U \approx 0.2\) in the lower right panel), all of which correspond to transitions involving a 2-electron occupancy. Approximation 4 (magenta stars), which includes 3-particle GFs at a mean-field limit, performs slightly better in this respect.

In Figure 3, we present results for the differential conductance obtained for the symmetric bridge where the inter-dot repulsion, \(V\), is included. All other parameter are identical to those of Figure 2. In this case, we find that approximation 1 is not suitable even for small values of the hopping term \(h\), while approximation 4 appears to work for low values of \(h < \frac{1}{2}U\) (both upper panels), however, it fails to capture peaks resulting from transitions through 2-electron occupancy at higher values of \(h\), as depicted in the lower panels of Figure 3. We
TABLE I. (Left column) Location of the poles of the unperturbed system’s GF as calculated using the 2nd approximation. (Right column) The differences in energy between many-particle states that diverge by one electron, such that $\Delta E(N) = E(N) - E(N - 1)$. Here $S_1 = \frac{1}{2}(U - V)^2 + 4h^2$, and $S_2 = \frac{1}{2}(U - V)^2 + 16h^2$.

| GF poles | Energy differences |
|----------|--------------------|
| $\pm |h|$ | $\Delta E(1)$: $\pm |h|$ |
| $\pm |h|$ | $\Delta E(2)$: $\pm |h|$ |
| $\pm (U + V)/2 - S_1 | + U + V - |h|$ |
| $\pm (U + V)/2 + S_1 | + U + V + |h|$ |

Note that for this set of parameters, such transitions involving 2 electrons are absent for $h < \frac{1}{2} U$ in the bias voltage studied. Approximations 2 and 3 agree very well with the ME results for all values of $h$, even at values of the bias voltage that correspond to transfer through 2-electron states, in contrast to the case where $V = 0$ in which they fail to capture conductance peaks involving 2 electrons.

The performance of the different approximations can be rationalized in terms of the pole structure of the unperturbed system GF, which can be compared to the exact many-body energy differences between many-particle states that diverge in one electron. While it is possible to carry out this analysis for all closure approximations, it is often a tedious task. Thus, in what follows we provide such an analysis for the case of approximation 2 only. The poles of the GF, the many-particle energy levels and the differences in energy are summarized in Table I (see the Appendix for more details regarding the derivation of the poles of the GF). As can be seen from Table I, conductance peaks corresponding to transitions $|0\rangle \rightarrow |1\rangle$ and $|1\rangle \rightarrow |0\rangle$, that is, peaks appearing at the values of $\Delta E(1)$, are captured by approximation 2 since the GF has poles at the correct locations. Higher excitations involving 2 or more electron occupancies are not fully or systematically accounted for by approximation 2 (or any of the other closures described in this paper, for this matter). In general, we find that such higher transitions are not captured by approximation 2 when $V \ll U$. For $V = U$ we find that $\Delta E(2)$ has 4 different values: $\epsilon + V \pm |h|$ and $\epsilon + V \pm 2|h|$, concurrently the GF has poles at $\epsilon + V \pm |h|$, thus, some of the transitions involving the 2-electron states $|N = 2\rangle$ (particularly those with $E(N = 2) = \epsilon + V \pm |h|$) are described by the NEGF.

Following this short analysis we can now better explain the results of Figure 3. For a large value of the inter-dot repulsion ($V = 0.8U$, thus $V \sim U$), one expects that the calculated GF will capture the higher order transitions in the relevant bias window and agree with the ME results. If one considers the second peak ($\Delta E \approx 1.1$) in the lower right panel of Figure 3, it results from transmission through a many-particle level with $|N = 2\rangle$. For the symmetric bridge, this peak corresponds to $\Delta E = (2\epsilon + \frac{1}{2}(U + V) - S_2) - (\epsilon - |h|) = \epsilon + \frac{1}{2}(U + V) - S_2 + |h|$, where $S_2 = \frac{1}{2}(U - V)^2 + 16h^2$. Under the assumption that $V \sim U$, one can approximate $\Delta E \approx \epsilon + V - |h|$, whereas the GF has one of its poles at $P_G = \epsilon + \frac{1}{2}(U + V) - S_1$, where $S_1 = \frac{1}{2}(U - V)^2 + 4h^2$, which, for $V \sim U$ can be approximated by $P_G \approx \epsilon + V - |h|$. For the studied parameters (see Figure 1) we find $P_G = 0.54289$ and $\Delta E = 0.54643$, and indeed the differential conductance based on approximation 2 shows a peak at twice this value $\Delta E \approx 1.1$. While for the case where $V = 0$ this transition is overlooked. It is easy to verify that a similar argument holds for the second peak in the lower left panel of Figure 3 as well.

B. Asymmetric bridge

We now turn to discuss the case where $\epsilon_{\text{sr}} \neq \epsilon_{\text{sl}}$ referred to as the asymmetric bridge. Once again we have calculated the differential conductance using the 4 different closure approximations to the NEGF formalism and compared the results to the differential conductance obtained by the ME. Analysis based on analytic expressions for the poles of the GF or the many-particle energies of $\hat{H}_3$ is more difficult, and the expressions are not as compact as in the symmetric case. The results for the poles of the GF within closure approximation 2 are given in the Appendix, while the many-body energy differences were obtained numerically.

In Figures 4 and 5, we plot the differential conductance for the asymmetric bridge for different values of the hopping term $h$ for $V = 0$ and $V = 0.8U$, respectively. The on-site single particle energies were $\epsilon_{\text{sr}} = \epsilon_{\text{sl}} = 0.15U$, $\epsilon_{\beta r} = \epsilon_{\beta l} = -0.2U$. The remaining model parameters are identical to those of the symmetric bridge and were taken to be $\Gamma_{L\alpha}^\uparrow = \Gamma_{L\alpha}^\downarrow = \Gamma_{R\beta}^\uparrow = \Gamma_{R\beta}^\downarrow = 0.015U$, $\Gamma_{L\beta}^\uparrow = \Gamma_{L\beta}^\downarrow = \Gamma_{R\alpha}^\uparrow = \Gamma_{R\alpha}^\downarrow = 0$, and $\beta^{-1} = U/40$. As before, the black curves (solid line) correspond to the ME results.

![FIG. 4. Plots of the differential conductance versus the bias voltage for the asymmetric bridge ($\epsilon_{\text{sr}} = \epsilon_{\text{sl}} = 0.15U$ and $\epsilon_{\beta r} = \epsilon_{\beta l} = -0.2U$) for $V = 0$. Upper left, upper right, lower left, and lower right panels correspond to $h = 0.1U$, $0.3U$, $0.5U$, and $0.7U$, respectively. Black curves correspond to results based on the ME. Red (circles), green (diamonds), blue (triangles), and magenta (stars) correspond to the results obtained by approximation schemes 1–4, respectively. The notation $|i\rangle \rightarrow |j\rangle$ indicates that the conductance peak calculated by means of ME corresponds to a transition from the $n_i$-particle states to any of the $n_j$-particle states. The remaining model parameters were $\Gamma_{L\alpha}^\uparrow = \Gamma_{L\alpha}^\downarrow = \Gamma_{R\beta}^\uparrow = \Gamma_{R\beta}^\downarrow = 0.015U$, $\Gamma_{L\beta}^\uparrow = \Gamma_{L\beta}^\downarrow = \Gamma_{R\alpha}^\uparrow = \Gamma_{R\alpha}^\downarrow = 0$, and $\beta^{-1} = U/40$.](image-url)
other curves represent the outcome of the NEGF formalism within the different closure approximations. We also label the different peaks in the differential conductance with the corresponding transitions between many-body states, i.e., \(|0\rangle \rightarrow |1\rangle\) corresponds to transitions from an empty system to a system with a single electron, etc.

From Figures 4 and 5, it is obvious that approximations 1 and 4 do not perform as well as approximations 2 and 3. We would like to note that approximations 1 and 4 utilized a mean-field like approximation decoupling the higher order GFs, while in approximations 2 and 3 higher order GFs are ignored altogether. For all parameters studied in this work (not all presented here), we find that approximation 2 performed better than all the other approximations, suggesting that including higher order correlations in a mean field fashion or a more complete treatment of the 2nd order GFs is not advantageous.

We find that approximations 2 and 3 predict negative differential conductance at higher values of \(h\), not obtained by the ME, as shown in the lower panels of Figure 5. The dips occur (in both cases) at values corresponding to the activation of the anti-bonding single electron state. While this transition is suppressed in the ME approach, it appears to be enhanced in the NEGF formalism.

IV. CONCLUDING REMARKS

In this work, we have assessed the validity of the EOM approach to the NEGF formalism for an interacting system coupled to two macroscopic leads. The interacting system consisted of two coupled quantum dots, each with one electronic level (spin up and spin down), connected serially, taking into account intra and inter-dot Coulomb interactions. Four different closure approximations to the EOM, some are commonly used in the literature and others have been developed here, were examined. As a measure of the quality of the approximations we calculated the differential conductance (derived by differentiating the steady state current with respect to the bias voltage) and compared the results to those obtained by the ME approach, which under the approximations of weak coupling to the leads and high temperature provides accurate results. Two different cases corresponding to a symmetric bridge \((\epsilon_{aa} = \epsilon_{ba})\) and an asymmetric bridge \((\epsilon_{aa} \neq \epsilon_{ba})\) with and without inter-dot Coulomb repulsion \((V)\), were studied for different values of the inter-dot hopping term \(h\). As expected, we find that keeping more terms in the closure or including higher order correlations in the EOM, does not necessarily improve the approximation. As a rule of thumb, neglecting higher order GFs (approximations 2 and 3) performs better as compared to closures that include such terms at a mean-field level (approximations 1 and 4).

To assess the performance of the different closure approximations, we compared the pole structure of the uncoupled GF with the exact results of the many-particle states. Focusing on approximation 2, which provides the overall best agreement in comparison to the ME approach, we found that transitions involving only single electron states were reproduced by the NEGF. However, when higher many-particle states are involved, the accuracy of the approximation depends on the strength of the Coulomb coupling \(V\). In cases where \(V \approx U\), approximation 2 also captures conductance peaks associated with transitions involving two electron states.

While all the approximations described in this work capture the Coulomb blockade and the main characteristics of the steady state transport for the single QD model (Anderson impurity model), they are not easily expandable to systems with more complex nonequilibrium dynamics, such as the double QD model (double Anderson model). In light of this, the success of approximation 2 for the double QD model does not necessarily imply that it will provide quantitative results for a system with more than a single level per a QD or for a system couplings more than two QDs. However, analysis of the poles of the resulting bare GF and comparing them to the exact many-body energy differences does provide a tool to assess the accuracy of a given approximate closure and can be used for larger bridge systems even if these can only be performed numerically.

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APPENDIX: POLE CALCULATION OF THE NEGF

Using the assumptions described in Sec. II B 2, the EOMs for the NEGF of the unperturbed system are (in Fourier space),

\[
C_{aa}^{\sigma\sigma} = (\hbar \omega - \epsilon_{aa})^{-1} \\
\times \left( 1 + hG_{\beta\alpha}^{\sigma\sigma} + U G_{\alpha\alpha}^{\sigma\sigma} + V G_{\beta\beta}^{\sigma\sigma} + V G_{\beta\alpha}^{\sigma\sigma} \right),
\]

\[
C_{\beta\alpha}^{\sigma\sigma} = (\hbar \omega - \epsilon_{\beta\alpha})^{-1} \\
\times \left( hG_{aa}^{\sigma\sigma} + U G_{\beta\beta}^{\sigma\sigma} + V G_{\alpha\alpha}^{\sigma\sigma} + V G_{\beta\alpha}^{\sigma\sigma} \right),
\]

\[
(A1)
\]
It is clear that the equations for the 2-particle GF close among themselves, so a simple substitution yields

\[
\begin{align*}
G_{\alpha\beta\sigma\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\beta}} - V \frac{h^2}{(\omega - \varepsilon_{\alpha\sigma} - U)}\right)^{-1} \\
G_{\alpha\sigma\alpha\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\sigma} - U} \frac{h}{\omega - \varepsilon_{\alpha\sigma}}\right)^{-1} \\
G_{\beta\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\beta\sigma} - V} \frac{h}{\omega - \varepsilon_{\beta\sigma}}\right)^{-1} \\
G_{\sigma\sigma\sigma\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\sigma\sigma} - V} \frac{h}{\omega - \varepsilon_{\sigma\sigma}}\right)^{-1} \\
G_{\alpha\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\sigma}} - V \frac{h^2}{(\omega - \varepsilon_{\alpha\beta} - U)}\right)^{-1} \\
G_{\alpha\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\sigma} - U} \frac{h}{\omega - \varepsilon_{\alpha\sigma}}\right)^{-1} \\
G_{\beta\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\beta\sigma} - V} \frac{h}{\omega - \varepsilon_{\beta\sigma}}\right)^{-1} \\
G_{\sigma\sigma\sigma\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\sigma\sigma} - V} \frac{h}{\omega - \varepsilon_{\sigma\sigma}}\right)^{-1} \\
G_{\alpha\beta\sigma\beta} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\beta}} - V \frac{h^2}{(\omega - \varepsilon_{\alpha\sigma} - U)}\right)^{-1} \\
G_{\alpha\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\sigma} - U} \frac{h}{\omega - \varepsilon_{\alpha\sigma}}\right)^{-1} \\
G_{\beta\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\beta\sigma} - V} \frac{h}{\omega - \varepsilon_{\beta\sigma}}\right)^{-1} \\
G_{\sigma\sigma\sigma\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\sigma\sigma} - V} \frac{h}{\omega - \varepsilon_{\sigma\sigma}}\right)^{-1} \\
G_{\alpha\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\sigma}} - V \frac{h^2}{(\omega - \varepsilon_{\alpha\beta} - U)}\right)^{-1} \\
G_{\alpha\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\sigma} - U} \frac{h}{\omega - \varepsilon_{\alpha\sigma}}\right)^{-1} \\
G_{\beta\sigma\beta\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\beta\sigma} - V} \frac{h}{\omega - \varepsilon_{\beta\sigma}}\right)^{-1} \\
G_{\sigma\sigma\sigma\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\sigma\sigma} - V} \frac{h}{\omega - \varepsilon_{\sigma\sigma}}\right)^{-1}.
\end{align*}
\]

We now substitute the set of equations (A6) into Eqs. (A5). Finally,

\[
\begin{align*}
G_{\alpha\beta\sigma\sigma} & = \left(\frac{h}{\omega - \varepsilon_{\alpha\beta}}\right)^{-1} \\
& \times \left(1 + \frac{hU_{\alpha\beta\sigma\beta}}{\omega - \varepsilon_{\alpha\beta}} + \frac{hV_{\alpha\alpha\sigma\sigma}}{\omega - \varepsilon_{\alpha\sigma}}\right) \\
& + \frac{hV_{\alpha\alpha\sigma\sigma}}{\omega - \varepsilon_{\alpha\sigma}} + U_{\alpha\alpha\sigma\sigma} + V_{\alpha\alpha\sigma\sigma} + V_{\alpha\beta\sigma\sigma}.
\end{align*}
\]

Define

\[
\begin{align*}
x_{\alpha\beta} & = \varepsilon_{\alpha\beta} / \varepsilon_{\alpha\beta}, \\
x_{\alpha\sigma\beta\sigma} & = \varepsilon_{\alpha\sigma} / \varepsilon_{\alpha\beta} + V, \\
x_{\alpha\sigma\beta\sigma} & = \varepsilon_{\alpha\sigma} / \varepsilon_{\alpha\beta} + U,
\end{align*}
\]

and rewrite the EOMs

\[
\begin{align*}
G_{\alpha\beta\sigma\sigma} & = \left(\frac{h}{\omega - x_{\alpha\beta}}\right)^{-1} \\
& \times \left(1 + hG_{\alpha\beta\sigma\sigma} + U_{\alpha\alpha\sigma\sigma} + V_{\alpha\alpha\sigma\sigma} + V_{\alpha\beta\sigma\sigma}\right),
\end{align*}
\]

\[
\begin{align*}
G_{\beta\sigma\beta\sigma} & = \left(\frac{h}{\omega - x_{\beta\sigma}}\right)^{-1} \\
& \times \left(hG_{\alpha\beta\sigma\sigma} + U_{\alpha\alpha\sigma\sigma} + V_{\alpha\alpha\sigma\sigma} + V_{\alpha\beta\sigma\sigma}\right).
\end{align*}
\]

From the last equation we see that the NEGF has poles at

\[
\begin{align*}
(\omega - x_{\alpha\beta})(\omega - x_{\beta\sigma}) - h^2 & = 0, \\
(\omega - x_{\alpha\sigma})(\omega - x_{\beta\sigma}) - h^2 & = 0, \\
(\omega - x_{\alpha\beta})(\omega - x_{\beta\sigma}) - h^2 & = 0,
\end{align*}
\]

\[
\begin{align*}
(\omega - x_{\alpha\sigma})(\omega - x_{\beta\sigma}) - h^2 & = 0, \\
(\omega - x_{\alpha\sigma})(\omega - x_{\beta\sigma}) - h^2 & = 0, \\
(\omega - x_{\alpha\sigma})(\omega - x_{\beta\sigma}) - h^2 & = 0,
\end{align*}
\]
or equivalently

\[
\begin{align*}
P_{G}^{1,2} &= \frac{1}{2}((x_a + x_p) \pm \sqrt{(x_a - x_p)^2 + 4h^2}], \\
P_{G}^{3,4} &= \frac{1}{2}((x_a + x_{fil}) \pm \sqrt{(x_a - x_{fil})^2 + 4h^2}], \\
P_{G}^{5,6} &= \frac{1}{2}((x_a + x_{flu}) \pm \sqrt{(x_a - x_{flu})^2 + 4h^2}], \\
P_{G}^{7,8} &= \frac{1}{2}((x_a + x_{fil}) \pm \sqrt{(x_a - x_{fil})^2 + 4h^2}].
\end{align*}
\]  

(A10)

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