Nanoscale device modeling using a conserving analytic continuation technique

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We propose an alternative approach to self-consistency and conservation laws in the theory of nonequilibrium Green's functions (NEGF's), which provides an infinite family of conserving but, generally, non-self-consistent approximations. Within any Φ-derivable approximation the associated Born series for the NEGF is shown to be conserving. Expectation values calculated from the Born series are then used to build a Padé table of approximations, while conservation laws are naturally preserved. We implement this technique for the Φ-derivable self-consistent Born approximation (SCBA), for which we obtain a recursion relation that yields the Born series for the NEGF up to any desired order. The expectation values calculated from the Born series are then postprocessed to build a Padé table of conserving approximations. The calculation of the SCBA photocurrent in a biased molecular junction model provides an example where, in addition to conservation laws, a substantial convergence acceleration relative to standard techniques is achieved. The present reformulation of the SCBA might aid convergence to the fully self-consistent results in a wide variety of problems.

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I. INTRODUCTION

Nonequilibrium Green's function (NEGF) techniques are commonly applied to the understanding and design of novel nanoelectronic devices.[1–4] In many situations one needs to take into account the interactions between electrons and other particles: interactions between electrons and phonons are a factor limiting the mobility in nanoelectronic devices;[5–8] for example, the photocurrent in a solar cell originates entirely from the interactions between electrons and photons and can be degraded or enhanced by electron-electron and electron-phonon interactions;[9] and electron-electron interactions give rise to electrostatic effects that modify charging energies of dopants and molecular junctions and to the Kondo and Coulomb-blockade effects which are difficult to account for.[10–13]

Thus approximate and inexpensive theoretical methods to treat interactions are, as always, very much in need. Unfortunately, due to lack of exact solutions and/or precise experiments, the accuracy associated with any chosen approximation is actually unknown. However, physical systems may have certain symmetries, and associated with these there are conserved quantities. Furthermore the physical laws used to model materials have also built-in symmetries, and thus associated conservation laws. For instance, the spherical symmetry of an atom guarantees the conservation of the total electronic angular momentum; the gauge symmetry in quantum electrodynamics guarantees charge conservation, i.e., the continuity equation and current conservation. When choosing an approximation to model material properties it is then paramount to ensure that the approximation respects these conservation laws to a good approximation. This is particularly important when the accuracy of the approximation remains unknown: a conserving approximation has, at least, the potential to produce physically meaningful results.

It is commonly believed that self-consistency is essential for an approximation to be conserving.[13] According to this view it is necessary to iterate complicated NEGF equations to self-consistency for an approximation to satisfy all necessary conservation laws. We shall see below that, strictly speaking, that is not the case. In fact, in Ref. 8 we have shown that full self-consistency is not a necessary condition for an approximation to be conserving (see also Ref. 7). Furthermore in Ref. 8 we have shown how, given a noninteracting expectation value of an observable, $\mathcal{O}_0$, one can evaluate a first-order correction, $\delta\mathcal{O}_1$, and then analytically continue the first-order expansion $\mathcal{O}_0 + \delta\mathcal{O}_1$ to obtain a new (conserving) approximation to the expectation value, as $\mathcal{O}_{AC} = \mathcal{O}_0/(1 - \delta\mathcal{O}_1/\mathcal{O}_0)$. In this work we ask, and answer in the affirmative, whether such technique can be generalized to all orders; the resulting approach produces an alternative iterative scheme where the Nth iteration produces $\sim N^2/2$ conserving approximations, some of which converge to the fully self-consistent results rather rapidly.

In the standard treatment of interactions within the NEGF formalism, one solves a Dyson equation for the interacting NEGF G,

$$G = g + g\Sigma[G]G, \tag{1}$$

where g is the noninteracting NEGF and Σ is a self-energy that accounts for the interactions, itself a functional of G and the interaction, $\Sigma = \Sigma[G]$. Thus one writes the fully interacting NEGF G as the noninteracting g plus a correction $g\Sigma[G]G$, which depends on itself through its dependence on G. Hence Eq. (1) must be, in principle, iterated to self-consistency. In Eq. (1) all quantities are time-dependent matrices in a given basis set and a convolution in a Keldysh-type contour is assumed for the time arguments.[1] According to Baym and Kadanoff[14] one can ensure conservation laws by means of Φ-derivable approximations for Σ, satisfying $\Sigma[G] = \delta\Phi[G]/\delta G$, where Φ is the Luttinger-Ward functional.[15] In practice the exact Φ is replaced by an approximation, and widely used approximations within condensed matter—such as the Hartree-Fock, self-consistent first and second Born, GW, and T-matrix approximations—are all Φ derivable.[14] Using a Φ-derivable approximation for Σ ensures that the observables calculated from G obey all conservation laws they ought to obey. For instance a poor Φ-derivable approximation yields unphysical electron densities and current densities

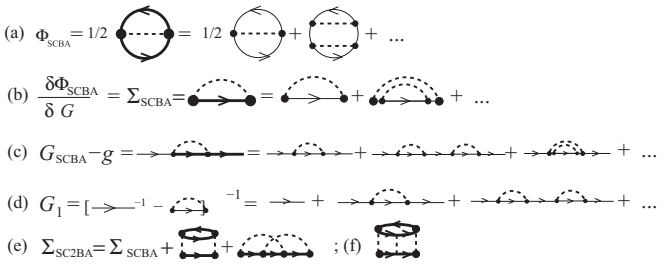

FIG. 1. Feynman diagrams corresponding to the SCBA (see text). Full lines are interacting NEGF's, thin lines noninteracting NEGF's, and dashed lines free boson GF's. (a) The SCBA Luttinger-Ward functional Φ. (b) The SCBA self-energy. (c) The SCBA NEGF. (d) G_1 calculated from Eq. (2) with $n = 1$. (e) Self-consistent second Born approximation, which includes second-order diagrams not accounted for by the SCBA. (f) A third-order diagram not accounted for by SC2BA. In (a)–(d) the rightmost equality shows the Born series to second order.

which are nevertheless related by the continuity equation. The Φ functional and Σ are illustrated in Figs. 1(a) and 1(b) for the self-consistent Born approximation (SCBA).

In practical calculations, Eq. (1) is written as $G = [g^{-1} - \Sigma[G]]^{-1}$ and then conventionally iterated in a computer by means of the expression

$$G_n = [g^{-1} - \Sigma[G_{n-1}]]^{-1}, \tag{2}$$

where $G_0 = g$. With G_0 one calculates $\Sigma[G_0]$, which is used to calculate G_1, and so on so forth until G_n and G_{n-1} agree within the specified tolerance.

There are some problems with this approach to Eq. (1): (*i*) For $n > 0$, the G_n's obtained on the way to self-consistency are not conserving; comparing Fig. 1(c) and Fig. 1(d) we see that G_1 reproduces the full self-consistent solution only to first order in the interaction, but not for higher orders; in G_1 these higher-order terms break Φ derivability—they miss, say, the rightmost diagram in Fig. 1(b)—and thus spoil the conservation laws. (*ii*) The sequence of G_n's given by Eq. (2) might converge slowly, if at all, to the fully self-consistent G; Eq. (2) then becomes a numerical burden. (*iii*) Self-consistent Φ-derivable approximations in practice always introduce errors because vertex corrections are not treated exactly:[16,17] as illustrated in Fig. 1 the SCBA NEGF is only exact to first order in the interaction [Fig. 1(c)], but not exact to second order [Fig. 1(e)]; the self-consistent second Born approximation [SC2BA, Fig. 1(e)] is exact to second order but not exact to third order, missing terms such as the one shown in Fig. 1(f). Then, as a result of (*iii*) above, self-consistent approximations typically wash out satellites in the spectral function.[13,16,17]

There is, thus, an important old dilemma: if one aims for conservation laws then one uses a Φ-derivable self-consistent approximation, in the process destroying important physical features in the spectral function. In this paper we show how one to avoid problems (*i*) and (*ii*). We introduce an infinite family of perfectly conserving non-self-consistent approximations, thus providing an alternative viewpoint on the topic of self-consistency and conservation laws in NEGF, challenging the conventional wisdom. The approach taken here might be particularly useful for the treatment of the sort of nonequilibrium problems that one encounters in nanoscale device modeling, but it is in fact a very general approach to estimate expectation values in quantum nonequilibrium systems.

The rest of the paper is organized as follows: in the next section we show that the Born series for the NEGF associated with any Φ-derivable approximation is conserving but divergent; in Sec. III we discuss the Padé technique used to "sum" divergent series, and how it might be used to estimate expectation values, while preserving conservation laws. In Sec. IV we provide an example of application, where the diagonal sequence of Padé approximants is used to calculate non-self-consistent conserved currents *and* to accelerate the convergence to the fully self-consistent ones. We end the paper with a discussion of the various implications of our results and the conclusions. For completeness we have included an Appendix on the Padé table.

II. CONSERVING, ORDER BY ORDER

In this section we show that the perturbation series for the NEGF, which we will also call Born series, is conserving for any order in the perturbation theory, provided that one uses a Φ-derivable approximation for the self-energy.

For any Φ-derivable approximation one can also build a Born series for the NEGF G as the partial series, g_N, involving powers of g and a coupling strength parameter U which multiplies the interaction:

$$g_N = g + \sum_{n=1}^{N} \delta g_n U^n = g + \sum_{n=1}^{N} \Delta g_n, \tag{3}$$

where the expansion coefficients $\delta g_n = \delta g_n[g]$ are U-independent functionals of g and the interaction, i.e., they depend on the particular functional form of the interaction but not on its strength, U. We have also defined $\Delta g_n = \delta g_n U^n$ for later use.

The Born series typically has a radius of convergence, U_c. For $U < U_c$ one then has $G = g_\infty$; by increasing N we can make g_N arbitrarily close to G, and thus conserving to arbitrary accuracy. But g_N is made of U-independent pieces, δg_n, and thus g_N is conserving independently of U, also for $U > U_c$. Thus we proved that the Born series is conserving for all U, when a Φ-derivable approximation is used. The arguments given by Baym work exactly as well when one replaces fully self-consistent G's by noninteracting NEGF g's, as Baym himself has pointed out.[18] For $U > U_c$, as is well known, one not only finds that $G \neq g_\infty$, but also that the conserving approximations g_N become worse and worse as N increases.

Independently of U, one can nevertheless evaluate the expectation value of an observable $\mathcal{O}$ from Eq. (3). If $\mathcal{O}$ is represented by a linear functional of G, satisfying $\mathcal{O}[g_1 + g_2] = \mathcal{O}[g_1] + \mathcal{O}[g_2]$, then one obtains a Born series for its expectation value,

$$\mathcal{O}_N = \mathcal{O}[g] + \sum_{n}^{N} o_n[\delta g_n] U^n, \tag{4}$$

where o_n is a functional of δg_n only. For $U < U_c$, increasing N increases the agreement between $\mathcal{O}_N$ and the fully self-consistent value $\mathcal{O}$. When the Born series diverges then the

sequence of approximates $\mathcal{O}_0 \equiv \mathcal{O}[g], \mathcal{O}_1, \mathcal{O}_2, \ldots$, consists of wildly different estimates for $\mathcal{O}$. The important message here is that if $\mathcal{O}$ is a conserved quantity then the estimates $\mathcal{O}_N$ are conserved, because they are calculated from g_N and g_N is conserving, independently of U. Once conservation laws are ensured the only thing left to do is to sum a divergent Born series for the expectation value.

III. SUMMING DIVERGENT SERIES WITH PADÉ

Fortunately in many cases divergent series can be summed and techniques to sum naturally have applications in all fields of physics.[19–23] Here we discuss one such technique, Padé analytic continuation, in relation to Φ derivability and conservation laws in nonequilibrium systems within the NEGF formalism. We have tried other techniques with good results—particularly the Borel technique,[19,22] the Shanks technique,[23] and Cesaro sums[19] —but they will be discussed elsewhere.[24,25]

Given a function $f(z)$ and its Taylor series $f_N(z) = \sum_{n=0}^{N} f_n z^n$, its $[l/m]$-Padé approximant is a rational function $f_{l/m}(z) = \mathcal{L}(z)/\mathcal{M}(z)$, where $\mathcal{L}(z)$ and $\mathcal{M}(z)$ are polynomials of degrees l and m with $l + m \leqslant N$ the coefficients of which are found from those of the Taylor series using well-known formulas, as shown in the Appendix.

Provided with the Taylor series to order $l + m$, one can then build a Padé table: the first row of this table $[l/0]$ gives the Taylor series to order l, $f_l(z) \equiv f_{l/0}(z)$; the second is Aitken's δ^2 method for convergence acceleration.[20] As pointed out by Cavassilas *et al.*[8] an important example is the function $f(z) = 1/(1+z)$, with Taylor series $f_{\infty/0}(z) = \sum_{i=0}^{\infty}(-z)^n$. For this function $f_{l/0}(z)$ is the Taylor or Born series to order l, which diverges for $|z| \geqslant 1$. One can check that the approximants $f_{l/l} = 1/(1+z)$, $\forall l$, all give the exact result.

Padé approximants are then our natural first choice to analytically continue expectation values calculated from g_N. Our procedure for calculating observables is then as follows: (*a*) Choose a Φ-derivable approximation. (*b*) Calculate the Born series for the Green's function as a partial sum, up to order N in the interaction, i.e., g_N. (*c*) From g_N calculate the Born series for the observable of interest up to order N, i.e., $\mathcal{O}_{N/0}$. (*d*) Use the partial sums $\mathcal{O}_{1/0}, \ldots, \mathcal{O}_{N/0}$ to build all possible Padé approximants and thus to analytically continue the Born series for the observable. Using this recipe one obtains a family of conserving approximations, a Padé table of $\mathcal{O}_{l/m}$'s.

To illustrate how the conserving property in the Born series is inherited by the Padé table let us consider as an example the continuity equation $\nabla J = -\partial\rho/\partial t$: we can evaluate both sides of this equation from g_N, which is conserving and therefore gives $\nabla J_{N/0} = -\partial\rho_{N/0}/\partial t$. With $\nabla J_{N/0}$ we can build a Padé table, which is of course the same Padé table that we obtain from $-\partial\rho_{N/0}/\partial t$. Therefore $\nabla J_{l/m} = -\partial\rho_{l/m}/\partial t$.

To summarize: the combination of Φ-derivable self-energy approximation with the associated conserving Born series for the NEGF and Padé analytical continuation for expectation values provides a way of building conserving approximations, which are not fully self-consistent. In the following sections we provide a full implementation of this idea at the level of the SCBA.

IV. THE SELF-CONSISTENT BORN APPROXIMATION

To implement this program we shall consider here the SCBA as a prototype, as it is the simplest nontrivial Φ-derivable approximation, and use it to produce new conserving approximations based on it. The SCBA is widely applied to model the effect of photons and phonons in nanoelectronics, and below we offer a fully operative approach that may be directly tried for that type of application. The SCBA self-energy is particularly simple, as it is linear in the electron NEGF, i.e., $\Sigma[g + \delta g] = \Sigma[g] + \Sigma[\delta g]$, and this fact can in turn be used to deduce a recursion relation that yields the Born series up to any order.

The recursion relation for the SCBA Born series to order U^N is obtained by substituting Eq. (3) on the right-hand side of Eq. (1), $g + g\Sigma[g_{N-1}]g_{N-1}$, and keeping terms up to order U^N and discarding any higher-order terms that may arise. Defining $\Delta g_0 = g$ and $\Sigma_N = \Sigma[\Delta g_{N-1}]$ one obtains the following formula to calculate Δg_N in Eq. (3):

$$g_N = g_{N-1} + g\sum_{n=0}^{N-1} \Sigma_{N-n}\Delta g_n\,, \tag{5}$$

which, in essence, is the recusion relation for the Catalan numbers.[24] Let us do a couple of iterations as an example. We start by setting $G = g$, evaluating $\Sigma_1 = \Sigma[g]$ to obtain Eq. (3) for $N = 1$, $g_1 = g + \Delta g_1$, with $\Delta g_1 = g\Sigma_1 g$. Now for $N = 2$, we define $\Sigma_2 = \Sigma[\Delta g_1]$, so that $\Sigma[g_1] = \Sigma_1 + \Sigma_2$. Next we evaluate $g(\Sigma_1 + \Sigma_2)g_1$ but keep terms up to U^2. This means discarding $g\Sigma_2\Delta g_1$ as it is $\propto U^3$. Then $g_2 = g + \Delta g_1 + \Delta g_2$ where $\Delta g_2 = g\Sigma_2 g + g\Sigma_1\Delta g_1$.

Note that g_N in Eq. (5) can also be obtained from G_N in Eq. (2): the Born series of G_N and g_N are both exact, and equal, up to order N; the difference is that G_N contains unphysical terms of order higher than N, while g_N does not by construction. For instance, to obtain g_1 one calculates G_1 for a rescaled interaction αU with $\alpha \ll 1$ so that $G_1 = g + \delta g_1 \alpha U$ holds. Then $\delta g_1 = (G_1 - g)/(\alpha U)$ and $\Delta g_1 = (G_1 - g)/\alpha$. Similar expressions may be easily found for $N > 1$. Thus in principle g_N can be calculated without change to existing numerical codes, based on Eq. (3).

Once we have g_N we can calculate observables from it, $\mathcal{O}_{N/0}$, and then build the table of $\mathcal{O}_{l/m}$'s with $l + m \leqslant N$.

V. EXAMPLE: PHOTOCURRENTS IN BIASED MOLECULAR JUNCTIONS

We apply this recipe to calculate the SCBA photocurrent in a biased molecular junction model at zero temperature. The model is chosen to provide a challenging test for the method put forward above, as well as an illustrative interesting application. To make it challenging we choose a model with the following properties: (*a*) The model contains a few states, to allow for multiple emission and absorption processes. (*b*) The states are narrow, with broadening $\sim\gamma$ and energy difference, $\Delta \gg \gamma$. (*c*) The relevant boson energy is Δ so that emission and absorption processes are resonant, and connect regions of high density of states. (*d*) There is no ballistic tunneling component, so that all tunneling processes are assisted by the emission and/or absorption of bosons and the analytic continuation formula proposed by Cavassilas *et al.*[8] does not apply. (*e*) The

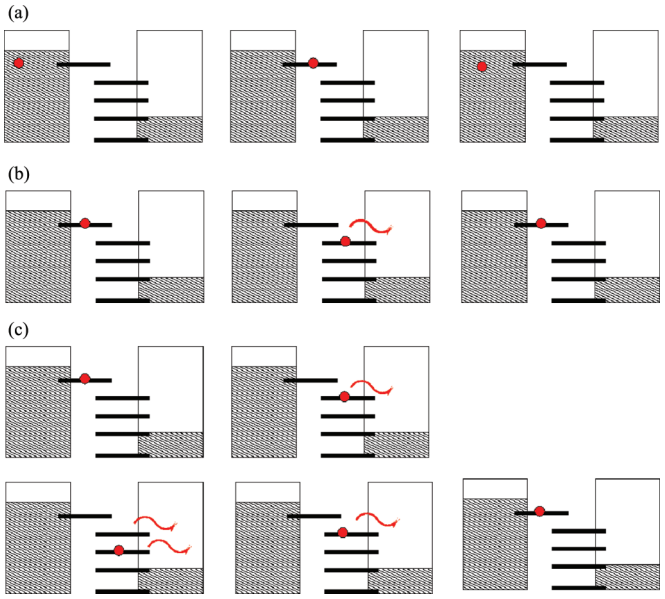

FIG. 2. (Color online) Photocurrent generation and degradation in a biased model molecular junction (see text). (a) In this model there is no direct tunneling. Electrons injected from the left electrode into the highest energy level are reflected back into the electrode. (b) One-photon processes generate a photocurrent: by emmiting and reabsorbing a photon electrons can make transitions between the highest level (coupled to the left electrode) and the second highest (coupled to the right electrode), thereby generating a photocurrent as tunneling into the right electrode becomes possible. (c) An example of two-photon process: we expect higher order photon processes to degrade the one-photon photocurrent, as the charge transported between L and R is now also involved in multiple transitions up and down the level ladder.

model contains an interaction strength parameter, enabling us to consider different interaction regimes, particularly those moderate interaction strengths at the limit of validity of the SCBA, where tunneling and scattering rates become comparable. For the sake of simplicity but without loss of generality we will neglect Hartree-type diagrams in our model. Thus the SCBA considered in this example includes only exchange-type diagrams.

Therefore we model the molecule (see Fig. 2 for the schematics) with five electronic states, corresponding to molecular orbitals (MO's) with equally spaced energies ϵ_i, $i = 0,4$, with $\epsilon_{i+1} = \epsilon_i + \Delta$. The molecule is coupled to left (L) and right (R) electrodes, with chemical potentials $\mu_{L/R}$. The highest MO, $i = 4$, couples only to the L electrode, while MO's 0–3 are coupled only to the R electrode. In the basis of MO's the noninteracting Hamiltonian, h_0, is diagonal, with matrix elements $(h_0)_{ij} = \epsilon_i \delta_{ij}$. The left and right electrode-coupling self-energies are $\Sigma^r_{L/R} = -i\Gamma_{L/R}/2$ and $\Sigma^a_{L/R} = (\Sigma^r_{L/R})^\dagger$, where $\Gamma_L = \gamma_L\,\mathrm{diag}(0,0,0,0,1)$ and $\Gamma_R = \gamma_R\,\mathrm{diag}(1,1,1,1,0)$ are the matrices describing the coupling between the MO's and the L and R electrodes, and $\gamma_{L/R}$ are the parameters that control the coupling strength and can differ. In the frequency domain, the lesser and greater electrode-coupling self-energies are $\Sigma^<_{L/R}(\omega) = i f_{L/R}(\omega)\Gamma_{L/R}$ and $\Sigma^>_{L/R}(\omega) = -i[1 - f_{L/R}(\omega)]\Gamma_{L/R}$, where $f_{L/R} = f_{L/R}(\omega;\mu_{L/R})$ are Fermi-Dirac distributions, parameterized by the L and R chemical potentials, $\mu_{L/R}$. In equilibrium $\mu_L = \mu_R = 0$, and out of equilibrium $\Delta\mu = \mu_L - \mu_R \neq 0$ is the applied bias voltage, which is applied symmetrically to both electrodes ($\mu_L = -\mu_R$). The noninteracting NEGF is given by $g^r(\omega) = [\omega I - h_0 - \Sigma^r_L - \Sigma^r_R]^{-1}$, $g^< = g^r(\Sigma_L + \Sigma_R)^< g^a$, $g^a = (g^r)^\dagger$, and $g^> = g^< + g^r - g^a$.

TABLE I. Summary of model parameters used for the molecular junction model discussed in the text. The parameters Δ, Ω, and $\gamma_{L/R}$ are fixed in the calculations shown below. The other parameters $\Delta\mu$ and M are varied in the ranges $\Delta\mu = 0$–1.5 V and $M = 0$–10 meV.

Δ (meV)	Ω (meV)	γ_L (meV)	γ_R (meV)	k_BT	μ
200	200	20	16	0	0

The units on this model are arbitrary and, for definitiveness, we choose a characteristic energy scale of a 100 meV, that is expected for molecules sandwiched between electrodes, where energy gaps are substantially renormalized by the dielectric environment; therefore all energies are in units of 0.1 eV (see Table I for a summary of relevant parameters). The MO with the highest energy is coupled to the L electrode and lower-energy MO's are all coupled to the R electrode. We choose $\Delta = 2$, so $h_0 = \mathrm{diag}(-7,-5,-3,-1,1)$, and $\gamma_L = 0.2$ and $\gamma_R = 0.16$.

In this noninteracting model there is no direct tunneling between the MO's. For $\Delta\mu = 0$, the highest MO is unoccupied, while lower-energy MO's are all occupied. For $\Delta\mu \geqslant \Delta$ the highest MO starts to become occupied, while the second-highest MO starts to become unoccupied. By increasing the bias lower energy states also become gradually unoccupied. Thus, as the bias is increased, phase-space is opened for transitions between states. Since there is no direct tunneling between MO's the ballistic current of the model is zero. The applied bias changes the electronic occupancies without generating a current. To produce a current we need to allow for an interaction which generates transitions between the L-coupled MO and the R-coupled MO's with lower energy. These transitions move charge between L and R electrodes, thereby generating a current, which we will refer to as a photocurrent.

The interaction is modeled after the electron-phonon (or -photon) interaction, described at the SCBA level. We consider the resonant case where there is only one boson mode of frequency $\Omega = \Delta$ described by a symmetric interaction matrix V, with matrix elements of the form $V_{i,j} = M\delta_{i,i\pm1}$ where M is the interaction strength parameter. This interaction matrix allows for resonant transitions between MO's with neighboring energies, separated by Δ, which thus generate a photocurrent. In the SCBA this interaction is accounted for by a self-energy $\Sigma^<(\omega)[G] = VG^<(\omega + \Delta)V$, $\Sigma^>(\omega)[G] = VG^>(\omega - \Delta)V$, and $\Sigma^r = (\Sigma^> - \Sigma^<)/2$; in the expression for $\Sigma^<$ the number of bosons is taken to be zero, so the electron liquid absorbs only bosons that have been emitted earlier. The interacting central region Green's function is calculated as $G^r = [\omega I - h_0 - \Sigma^r_L - \Sigma^r_R - \Sigma^r]^{-1}$ and $G^< = G^r(\Sigma^<_L + \Sigma^<_R + \Sigma^<)G^a$. We then iterate these equations to self-consistency, which is equivalent to Eq. (2) within the SCBA. With the self-consistent G we calculate

the photocurrent at the interface between electrode $\alpha = L/R$ and the molecule $\mathcal{I}^{\alpha}[G] = e/h \int d\omega \,\mathrm{Tr}[G^{>}\Sigma_{\alpha}^{<} - G^{<}\Sigma_{\alpha}^{>}]$. In the steady state, current conservation implies $I^{L} = -I^{R}$. Alternatively, from Eq. (5) one can also calculate g_N and from it $\mathcal{I}^{\alpha}[g_N] = \mathcal{I}^{\alpha}_{N/0}$; to do so we use Langreth rules in terms of the form $(g\Sigma_{N-i}g_i)^{r,<}$ and calculate $\Delta g_N^{<,r}$. Since in the Born series for G every order is conserving, $\mathcal{I}^{L}_{N/0} = -\mathcal{I}^{R}_{N/0}$. Then sequences of estimates $\mathcal{I}^{L}_{N/0}$ and $-\mathcal{I}^{R}_{N/0}$ are identical and have thus the same Padé table. We will restrict ourselves to the diagonal sequence of this table, which satisfies then $\mathcal{I}^{L}_{l/l} = -\mathcal{I}^{R}_{l/l}$, $2l \leqslant N$.

In our calculations we obtain numerically perfect satisfaction of the current conservation law $\mathcal{I}^{L}_{N/0} = -\mathcal{I}^{R}_{N/0}$, and thus $\mathcal{I}^{L}_{l/l} = -\mathcal{I}^{R}_{l/l}$. This observation remains true for any combination of model parameters we tried, and for the various other models we have considered.[8,24,25] In our SCBA calculations current is also conserved to a good approximation; our convergence criterion ensures violations to be smaller than 0.1%.

In Fig. 3 we plot the photocurrent as a function of applied bias. The top panel shows a comparison between the Born photocurrents $\mathcal{I}_{N/0}$ and the fully self-consistent SCBA ones, for various values of M and $N = 1,3,5$. For $\Delta\mu < \Delta$ there is no phase-space for transitions and the photocurrent is zero. For $\Delta\mu \sim \Delta$ transitions between levels 4 and 3 become possible; for $\Delta\mu > 3\Delta$, $3 \leftrightarrow 2$ transitions are allowed and so on. Therefore as the voltage is increased new regions of phase-space become available abruptly at $\Delta\mu = \Delta, 3\Delta, 5\Delta$, and 7Δ and one observes steps in the photocurrent at these values of the bias voltage.

When the electron-boson interaction M is small, one-boson processes set up a photocurrent which grows as M^2; in this limit all approximations agree. As M increases, see Fig. 3(a), two- and three-boson processes become important, and have the effect of degrading the one-boson photocurrent, $\mathcal{I}_{1/0}$. The Born series converges and the estimates $\mathcal{I}_{N/0}$ become better as we increase N. Unfortunately in Figs. 3(b) and 3(c) we see that just by doubling the value of M the Born series diverges, and increasing N and/or M just makes things worse.

In the bottom panels, Figs. 3(d)–3(f), we compare the SCBA with the diagonal elements of the Padé table, $\mathcal{I}_{L/L}$, $L = 1,2,3$. The evaluation of $\mathcal{I}_{L/L}$ requires $2L$ iterations of Eq. (5). The agreement with the full SCBA results is excellent, even for $M \sim \gamma_{L/R}$, well outside the radius of convergence for the Born series and close to the limit of validity of the SCBA. The approximation $\mathcal{I}_{1/1}$ is as complicated as $\mathcal{I}_{2/0}$, but it provides a fairly good approximation to the SCBA data, even for quite large values M. Both approximations $\mathcal{I}_{2/2}$ and $\mathcal{I}_{3/3}$ essentially agree with the SCBA current, exhibiting the rapid convergence of the diagonal Padé sequence to the SCBA data. A simplistic picture of the physics of this model can be as follows: one-boson processes generate the photocurrent; processes involving more than one boson tend to degrade the one-boson photocurrent. Then we define a high-bias photocurrent-degradation parameter $\mathcal{D} = 100(1 - \mathcal{I}/\mathcal{I}_{1/0})$. From the data shown in Fig. 2(f) we obtain $\mathcal{D}_{SCBA} = 69\%$, while $\mathcal{D}_{1/1} = 79\%$, $\mathcal{D}_{2/2} = 72\%$, and $\mathcal{D}_{3/3} = 70\%$. We would like to point out that $\mathcal{I}_{1/1}$, $\mathcal{I}_{2/2}$, and $\mathcal{I}_{3/3}$ were calculated in 2, 4, and 6 iterations of Eq. (5), respectively. We find it remarkable that in the present model the 1/1 Padé approximant is able to account for large photocurrent degradations in just two iterations of Eq. (5), capturing 70% of the SCBA photocurrent at large bias, for $M = 10$ meV ($\sim\gamma$).

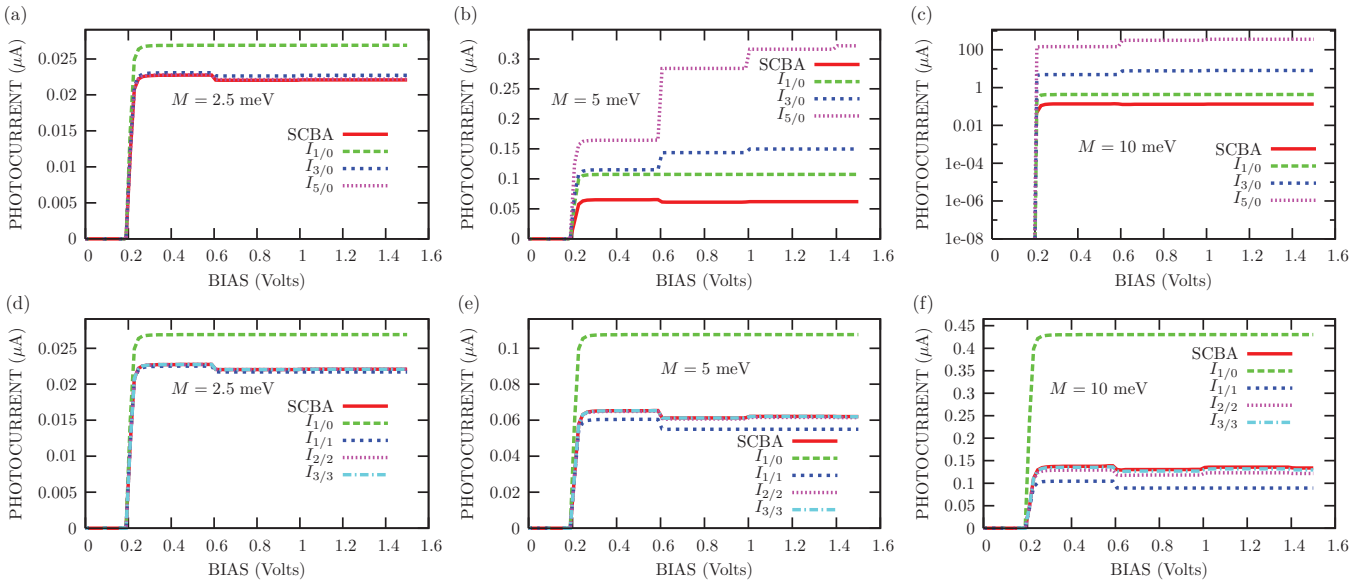

FIG. 3. (Color online) Photocurrent (in μA) as a function of source-drain bias (in volts). In (a)–(c) we show a comparison between currents calculated from the SCBA and the Born series ($I_{N/0}$), for $N = 1$, 3, and 5, and the values of M shown. (a) For $M = 2.5$ meV excellent agreement between SCBA and Born series currents are obtained, already for $N = 3$. (b),(c) For $M = 5$–10 meV, the Born series is already divergent, blowing up badly as M is increased—note the log scale in (c). In (d)–(f) we compare SCBA currents with those obtained from the Padé analytic continuation of the Born series for the current, $I_{1/1}$, $I_{2/2}$, and $I_{3/3}$. The Padé approximant $I_{3/3}$ reproduces the exact SCBA result, and is calculated from the Born series including terms up to sixth order. For $M = 10$ meV and high bias, $I_{1/1}/I_{\mathrm{SCBA}} = 0.68$, $I_{2/2}/I_{\mathrm{SCBA}} = 0.90$, and $I_{3/3}/I_{\mathrm{SCBA}} = 0.97$.

VI. DISCUSSION

In this work we have proposed a different take on selfconsistency and conservation laws. Rather than iterating Dyson's equation in the usual way, one can start from a Φ-derivable self-energy approximation and obtain the Born series for the NEGF, which we have shown to be conserving, and from it one calculates a series for the expectation value. When the series diverges or converges slowly, analytic continuation techniques offer new conserving approximations and convergence acceleration.

The technique put forward in this work offers several distinct advantages over more widely followed routes to self-consistency and conservation laws in NEGF's. We would like to point out the following advantages: (*i*) Recursion relations to calculate the Born series, such as Eq. (5), involve only matrix products, and no matrix inversion, which is less accurate. (*ii*) By calculating expectation values from g_N, $\mathcal{O}_{N/0}$, one obtains also $\sim N^2/2$ perfectly conserving approximations—the Padé table. This is to be compared with the standard technique, producing only one conserving approximation, the fully self-consistent one. (*iii*) When M is very large one might find difficulties converging Eq. (2) and the Padé table can accelerate the convergence towards the fully self-consistent results.

Our results can be seen as a generalization of our previous work at the lowest order.[8] Mera *et al*. showed how conservation laws are obeyed by a lowest-order approximation. Here the same has been shown for any order. Based purely on intuitive grounds Cavassilas *et al*. proposed using the zero-order current and first-order correction $\mathcal{I}_0$ and $\delta\mathcal{I}_1$ to build the approximation $\mathcal{I}_0/(1-\delta\mathcal{I}_1/\mathcal{I}_0)$, which we now identify as the [0/1] Padé approximant to the current, $\mathcal{I}_{0/1}$. So the simplest conserving one-iteration approximation calculate the ballistic $\mathcal{I}_0$, the first order $\mathcal{I}_{1/0}$, and the first Padé approximant $\mathcal{I}_{0/1}$.

As presented here the technique is suitable only for electron-boson interactions. The case of electron-electron interactions is more challenging, as one should start from fully self-consistent Hartree-Fock result—or similar approximations, as noted by Steiner *et al*. and confirmed by calculations performed by one of us.[26,27] The reason for this is that in the case of electron-electron interactions there is typically a large quasiparticle shift that needs to be accounted for to infinite order. Then one can evaluate the second-order self-energy, shown in Fig. 1(e) using the fully self-consistent Hartree-Fock Green's function, i.e., a second order correction, which can then be used to compute expectation values and one Padé approximants, which then approximates well the fully self-consistent second-order results for electron-electron interaction strengths from weak to moderate.

Therefore the accuracy of the Padé technique depends on the system under consideration. It is only one of the analytic continuation methods and perhaps not always the best. However, we find it to be physically appealing, conserving and, in the present calculations and those of Ref. 8, as accurate as the fully self-consistent approximation. We are actively investigating the limits of validity of the method, as well as alternative analytic continuation strategies.[24,25]

VII. CONCLUSIONS

In conclusion we have considered an alternative route to self-consistency and conservation laws within the NEGF formalism. Using a Φ-derivable self-energy the resulting Born series for the NEGF is conserving. One then calculates expectation values from the Born series and uses Padé analytic continuation to obtain new approximations to the expectation value. These expectation values then satisfy whatever conservation laws they ought to satisfy. In addition, in the example given, great convergence acceleration to the fully self-consistent results has been achieved. Therefore we have tackled the problems (*i*)–(*ii*) that we mentioned in the introduction. We have not addressed in this work the more challenging problem (*iii*), vertex corrections. Perhaps the techniques put forward in this paper may help one day advance us towards the solution of that problem.

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APPENDIX: THE PADÉ TABLE

1. Building the Padé table

For the sake of completeness, in this Appendix we provide a more in-depth discussion on the Padé table. For more details see Ref. 20; for an excellent Appendix on the numerical evaluation of Padé approximants see Ref. 21. Given an arbitrary function $f(z)$ and its Taylor series to order N, $f_N(z)=\sum_{n=0}^{N} f_n z^n$, we wish to calculate a set of rational approximations $f_{l/m}(z)=\mathcal{L}(z)/\mathcal{M}(z)$, where $\mathcal{L}(z)=\mathcal{L}_0+\mathcal{L}_1 z+\mathcal{L}_2 z^2+\cdots+\mathcal{L}_l z^l$ and $\mathcal{M}(z)=1+\mathcal{M}_1 z+\mathcal{M}_2 z^2+\cdots+\mathcal{M}_m z^m$ are polynomials of degrees l and m with $l+m\leqslant N$. The Taylor series of $f_{l/m}(z)$ and $f(z)$ are then identical to order $l+m$. To find the coefficients $\mathcal{L}_i$ and $\mathcal{M}_i$ from those of the Taylor series we use the following wellknown formulas:[20]

$$\mathcal{L}_0 = f_0, \tag{A1}$$

$$\mathcal{L}_1 = f_1+f_0\mathcal{M}_1, \tag{A2}$$

$$\mathcal{L}_2 = f_2+f_1\mathcal{M}_1+f_0\mathcal{M}_2, \tag{A3}$$

$$\cdots$$

These equations are closed using the fact that $\mathcal{L}_x=0$ for $x>l$ and $\mathcal{M}_y=0$ for $y>m$. For an arbitrary function $f(z)$, the resulting rational approximations can be arranged into a table—the Padé table. For instance, given the Taylor series of $f(z)$ to second order, one obtains six approximations to $f(z)$,

$$f_{l/m}=\begin{pmatrix} f_0 & f_0+f_1z & f_0+f_1z+f_2z^2 \\ \frac{f_0}{1-f_1z/f_0} & \frac{f_0+(f_1-f_0f_2/f_1)z}{1-f_2z/f_1} & \\ \frac{f_0}{1-f_1z/f_0+((f_1/f_0)^2-f_2/f_0)z^2} & & \end{pmatrix}=\begin{pmatrix} f_{0/0} & f_{1/0} & f_{2/0} \\ f_{0/1} & f_{1/1} & \\ f_{0/2} & & \end{pmatrix}. \tag{A4}$$

2. Two examples

Let us then evaluate the second-order Padé table for a couple of functions. First we consider the function $f(z) = 1/(1+z)$, which is slowly varying but its Taylor series has a finite radius of convergence (=1). For this function $f_n = (-1)^n$, and its second order Padé table is then

$$f_{l/m} = \begin{pmatrix} 1 & 1-z & 1-z+z^2 \\ \frac{1}{1+z} & \frac{1}{1+z} & \\ \frac{1}{1+z} & & \end{pmatrix}, \tag{A5}$$

and the Padé table reproduces the exact result, except for its first row which gives back the Taylor series.

To check the extent to which this promise may materialize one considers instead a more rapidly varying function, $f(z) = e^{-z}$, with a Taylor series which converges everywhere and expansion coefficients to second order given by $f_0 = 1$, $f_1 = -1$, and $f_2 = 1/2$. The second-order Padé table is then

$$f_{l/m} = \begin{pmatrix} 1 & 1-z & 1-z+z^2/2 \\ \frac{1}{1+z} & \frac{1-z/2}{1+z/2} & \\ \frac{1}{1+z+z^2/2} & & \end{pmatrix}, \tag{A6}$$

$$\mathcal{I}_{L/M} = \begin{pmatrix} \mathcal{I}_0 & \mathcal{I}_0+\Delta\mathcal{I}_1 & \mathcal{I}_0+\Delta\mathcal{I}_1+\Delta\mathcal{I}_2 \\ \frac{\mathcal{I}_0}{1-\Delta\mathcal{I}_1/\mathcal{I}_0} & \frac{\mathcal{I}_0+\Delta\mathcal{I}_1-\mathcal{I}_0\Delta\mathcal{I}_2/\Delta\mathcal{I}_1}{1-\Delta\mathcal{I}_2/\Delta\mathcal{I}_1} & \\ \frac{\mathcal{I}_0}{1-\Delta\mathcal{I}_1/\mathcal{I}_0-\Delta\mathcal{I}_2/\mathcal{I}_0+(\Delta\mathcal{I}_1/\mathcal{I}_0)^2} & & \end{pmatrix}. \tag{A7}$$

Here we recognize the lowest-order approximation (first order for the SCBA) discussed by Mera *et al.*,[8] $\mathcal{I}_{1/0} = \mathcal{I}_0 + \Delta\mathcal{I}_1$. The LOA+AC method introduced by Cavassilas *et al.*[8] is the 0/1 Padé approximant: $\mathcal{I}_{0/1} = \mathcal{I}_0/(1 - \Delta\mathcal{I}_1/\mathcal{I}_0)$. We would like to point out that the example discussed in the text has no ballistic current, $\mathcal{I}_0 = 0$, and therefore the first column in the Padé table does not give meaningful results. Thus the analytic continuation formula given by Cavassilas *et al*. does not apply in the example discussed here. In those cases the table should start in the first column; the first nontrivial Padé approximants are of second order and are obtained from the table by setting $\mathcal{I}_0 = 0$.

which, at first sight, does not look as promising. To see the convergence acceleration at work one can for instance expand $f_{1/1}(z)$ to fourth order, $f_{1/1}(z) = (1 - z/2)/(1 + z/2) = 1 - z + z^2/2 - z^3/4 + 3z^4/16 + \mathcal{O}(z^5)$, and consider the difference $e^{-z} - f_{11}(z) = z^3/12 - 7z^4/48 + \mathcal{O}(z^5)$. Compared to e^{-z}, the fourth-order difference, $z^3/12 - 7z^4/48$, is very small in the interval $z \in [0,1]$. So for $f(z) = e^{-z}$ and $z \in [0,1]$, the 1/1 Padé approximant is a second-order approximation with roughly fourth-order accuracy.

3. Padé table for the current

Finally we give the explicit Padé table for the current up to second order. The expressions for higher orders become too long and complicated, and we recommend to evaluate them numerically instead.[21] Imagine that one calculates the ballistic current $\mathcal{I}_0$ and the first- and second-order corrections, $\Delta\mathcal{I}_1$ and $\Delta\mathcal{I}_2$, which are proportional to U and U^2 respectively. Then one writes down the following Padé table for the current:

4. Limitations of the Padé table

Finally, we would like to point out that in any given rational approximation the denominator can vanish, and therefore particular entries in the Padé table may diverge for specific choices of parameters. By increasing the order in the Padé table this divergences appear at increasingly higher values of the interaction strength parameter, and the approximation is therefore typically improved. We also note that different rows, columns, and "paths" within the table have different convergence properties.[20] According to Padé folklore, the diagonal sequence is the best.[20]

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