Nonequilibrium coherent potential approximation for electron transport

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Accounting for the effects of disorder on the transport properties of electronic devices is indispensable for comparison with experiment. However, theoretical treatment of disorder presents essential difficulty because the disorder breaks the periodicity of the system. The coherent potential approximation (CPA) solves this problem by replacing the disordered medium with a periodic effective medium. However, calculating the electron current within CPA requires summing scattering diagrams to infinite order called vertex corrections. In this work we reformulate CPA for nonequilibrium electron transport. This approach, based on the nonequilibrium Green's function formalism, greatly simplifies the treatment of disordered transport by eliminating the vertex corrections.

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I. INTRODUCTION

Disorder is present in electronic devices either by design (e.g., doping) or as an inevitable side effect of the experimental process. Therefore, for theoretical models to quantitatively compare with experiments, it is necessary that the disorder be included in the models. Unfortunately, in the present state of theory, accounting for the effects of disorder on the transport properties of devices is too cumbersome, and it is rarely done in practice. The vast majority of theoretical reports involve ideal systems. The reason is that calculating the electronic structure and properties relies essentially on the periodicity of the system. Thus, there is a principal difficulty in treating random disorder because it breaks this periodicity.

There are many possible types of disorder but the most commonly considered are (i) substitutional disorder when impurity atoms substitute randomly for host atoms in the lattice and (ii) magnetic disorder when the magnetic moment directions deviate randomly from the spin quantization axis. The coherent potential approximation (CPA) is a powerful tool to threat these types of disorder.[1] It is based on the the Green's function (GF) formalism, which is especially suitable for that purpose because a perturbative expansion can be developed for the GF.[2] CPA replaces the actual environment seen by an electron with an effective periodic medium which is characterized by a complex self-energy. This self-energy or energy-dependent coherent potential is obtained self-consistently from the requirement that the scattering vanishes on average in the effective medium. Originally CPA was introduced by Soven in 1967[3] and independently by Taylor.[4] Velicky *et al*. formulated the single-site approximation to the CPA.[5]

The CPA technique for electron transport was developed by Velicky in 1969 for the purpose of calculating the conductivity in disordered systems using the Kubo formula.[6] The steady-state electron current of a noninteracting system can be expressed through the product of the retarded and advanced GFs.[7,8] In the presence of disorder the GFs cannot be decoupled and the current expression contains diagrams to infinite order. The first term, called the "bubble" represents the specular conductance. The rest of the terms, known as the vertex corrections (VCs), give all the contributions to the diffusive conductance. Comparing CPA with brute force supercell calculations shows that taking into account the diffusive contributions is essential to describe the conductance correctly.[9–11] Therefore, VCs are extremely important; however, calculating them can present a substantial technical difficulty.[12] Recently, it was shown that the on-site vertex function which appears within the single-site CPA formalism plays the role of the local chemical potential within Büttiker's voltage-probe approach.[13]

CPA has been implemented within the framework of several electronic structure methods, such as Korringa-Kohn-Rostoker (KKR)[14,15] and linear muffin-tin orbitals (LMTO).[16,17] Recently Guo *et al*. showed that calculating steady-state current using the charge density from first-principles calculations can lead to large errors.[18] They introduces a "nonequilibrium vertex correction" (NVC) to account for the nonequilibrium charge distribution. However, this NVC is a correction to the electronic structure, not the current, which is calculated using the standard CPA plus VCs methodology.

In this work we propose a nonequilibrium formulation of CPA (NE-CPA) for electron transport through disordered systems. The method is based on the nonequilibrium GF (NEGF) formalism.[19] Within this formalism there are two independent GFs, the retarded G and the Keldysh F. The proposed method relies on the fact that the nonequilibrium current operator depends linearly on the Keldysh GF. Therefore, in the presence of disorder the current is directly expressed through the NEGF of the effective medium. Within NE-CPA the expression for the current contains only one term which is equivalent to CPA plus VCs to infinite order. In addition the Keldysh GF already contains the nonequilibrium charge distribution information. Thus NE-CPA provides a much simpler, precise, and efficient method to calculate transport in disordered systems.

II. METHODOLOGY

We assume that the wave function can be expanded in terms of a linear combination of orbitals localized around each atomic site (LCAO). In this basis the Hamiltonian of the system can be written as

$$H = \sum_{nm,\alpha\beta,\sigma} H^{\sigma}_{nm,\alpha\beta} c^{\dagger}_{n\alpha\sigma} c_{m\beta\sigma}, \tag{1}$$

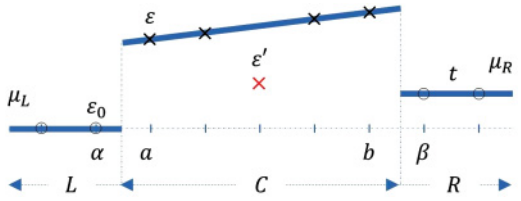

FIG. 1. (Color online) Schematic view of the two-probe setup consisting of two electrodes and a scattering region. In this case, the scattering region is a tunnel barrier with a disordered layer in the middle.

where n,m are site, α,β orbital, and σ spin indices. The H^{σ}_{nm} is the Hamiltonian matrix element between two sites n and m for spin σ. The current is normally calculated in the two-probe setup, schematically shown in Fig. 1. The current flows between the left (L) and right (R) electrodes through a central scattering region (C).

The continuity equation requires that the current should be the same anywhere in the system. Therefore, the current operator is obtained from the continuity condition to be

$$\hat{I} = \frac{e}{i\hbar}\sum_{n,\alpha\beta,\sigma}(H_{ln,\alpha\beta,\sigma}c^{\dagger}_{l\alpha\sigma}c_{n\beta\sigma} - H_{nl,\alpha\beta,\sigma}c^{\dagger}_{n\alpha\sigma}c_{l\beta\sigma}). \quad (2)$$

We adopt the principal layer (PL) convention according to which the PL is the smallest unit cell in the z direction such that there is hopping only between nearest-neighbor PLs. Therefore, the site position is $\mathbf{r} = \mathbf{R} + \tau$ where $\mathbf{R} = (\rho,z)$ is a lattice vector consisting of the two-dimensional vector ρ in the plane and the PL number z and τ is the atomic positions within the PL. Thus, the site index can be decomposed to $n = (\rho,z,\tau)$. Then the expectation value of the operator gives the physical current through PL i to be[7]

$$I = \frac{e}{\hbar}\sum_{\rho\rho'}\mathrm{Tr}[H_{i+1,i}(\rho',\rho)F_{i,i+1}(\rho,\rho';t,t_+) - H_{i,i+1}(\rho',\rho)F_{i+1,i}(\rho,\rho';t,t_+)], \quad (3)$$

where both H and F are matrices with respect to the indices (τ,α,σ) and matrix multiplication is assumed. The $F = G^{<} + G^{>}$ is the Keldysh NEGF.[19]

The expression above is general and applies to any system out of equilibrium. In the steady-state situation, we can make a Fourier transform from time to energy space

$$I = \frac{e}{\hbar}\int dE\ \mathrm{Tr}[H_{i+1,i}F_{i,i+1}(E) - H_{i,i+1}F_{i+1,i}(E)], \quad (4)$$

where now the trace includes a sum over all repeating indices including the in-plane vectors. This formula has the simple interpretation as the net flow of charge through a surface between PL i and $i+1$. Due to the continuity equation, the PL i can be any PL, either in the electrodes or in the scattering region. Customarily it is chosen to be at the interface.

The Hamiltonian of the two-probe system without disorder can be decomposed as

$$H = H_C + H_L + H_R + (H_{CL} + H_{CR} + \mathrm{H.c.}), \quad (5)$$

where H_C, H_L, and H_R are the Hamiltonian matrix elements of the isolated scattering region and left and right electrodes; $H_{CR(CL)}$ is the coupling between the scattering region and the electrodes. We can derive expressions for the retarded and the Keldysh GF under the assumption that each electrode of the system is maintained at a local equilibrium with two different chemical potentials μ_L and μ_R. As input we have the GF of the disconnected parts, g_{CC} and $g_{LL(RR)}$. The GF of the isolated scattering region is $g_{CC} = (E - H_C)^{-1}$. The surface GFs for the semi-infinite electrodes are obtained using one of the available standard methods.[20,21] Using those we can construct the "connected" retarded GF of all regions by treating the overlap between them as perturbation $V = H_{CL} + H_{CR} +$ H.c. Projecting the Dyson equation[2] on the central region we get

$$G^0_{CC} = g_{CC} + g_{CC}H_{CL}G^0_{LC} + g_{CC}H_{CR}G^0_{RC}, \quad (6)$$

where G^0_{CC} is the retarded GF without disorder of the scattering region coupled to the leads and $G^0_{L(R)C}$ is the retarded GF between the left (right) lead and the scattering region. Then we project the Dyson equation at the interface to obtain

$$G^0_{L(R)C} = g_{LL(RR)}H_{L(R)C}G^0_{CC}. \quad (7)$$

Then, combining these two equations yields

$$G^0_{CC} = g_{CC} + g_{CC}\Sigma G^0_{CC}, \quad (8)$$

where $\Sigma = \Sigma_L + \Sigma_R$ is the self-energy due to connection of the scattering region to the electrodes

$$\Sigma_{L(R)} = H^{\dagger}_{L(R)C}g_{LL(RR)}H_{L(R)C}. \quad (9)$$

Note that the self-energy matrix elements are in the scattering region. Here we use the zero superscript to signify the quantities before disorder is introduced. Thus, the expression for the connected GF becomes

$$G^0 = (I - g\Sigma)^{-1}g = (E - H_C - \Sigma_L - \Sigma_R)^{-1}. \quad (10)$$

Next we write down the quantum kinetic equation[19] for the "connected" Keldysh GF projected on the scattering region without disorder

$$F^0_{CC} = f^0_{CC} + f^0_{CC}H_{CL}G^{0\dagger}_{LC} + g_{CC}H_{CL}F^0_{LC} + f^0_{CC}H_{CR}G^{0\dagger}_{RC} + g_{CC}H_{CR}F^0_{RC}, \quad (11)$$

where f^0_{CC} is the Keldysh GF in the isolated scattering region and should not be confused with the Fermi-Dirac distribution function f. Caroli *et al.* assumed that the retarded and advanced GFs of the isolated barrier are real yielding that the Keldysh GF of the isolated barrier is zero.[7] It can be shown in general that even if there are states in the scattering region and the Keldysh GF of the isolated scattering region is nonzero, the steady-state current does not depend on the Keldysh GF of the isolated scattering region and the terms proportional to f^0 can be dropped out:[22]

$$F^0_{CC} = g_{CC}H_{CL}F^0_{LC} + g_{CC}H_{CR}F^0_{RC}. \quad (12)$$

Next we project the quantum kinetic equation on the interface:

$$F^0_{L(R)C} = f^0_{LL(RR)}H_{L(R)C}G^{0\dagger}_{CC} + g_{LL(RR)}H_{L(R)C}F^0_{CC}, \quad (13)$$

where $f^0_{LL(RR)} = (2f_{L(R)} - 1)(g^{\dagger}_{LL(RR)} - g_{LL(RR)})$ is the Keldysh GF of the isolated left (right) lead which are assumed in equilibrium. Combining these two equations yields

$$F^0_{CC} = iG^0_{CC}[(2f_L - 1)\Gamma_L + (2f_R - 1)\Gamma_R]G^{0\dagger}_{CC}, \quad (14)$$

where $\Gamma_{L(R)}$ is proportional to the imaginary part of the self-energy $\Sigma_{L(R)}$

$$\Gamma_{L(R)} = i(\Sigma_{L(R)} - \Sigma^{\dagger}_{L(R)}) \tag{15}$$

and has the meaning of the lifetime of the states in the central region (finite due to the connection to the electrodes) or in other words it is the escape rate to the leads. Furthermore, substituting this in the expression for the current Eq. (4), we obtain that under steady-state conditions the current can be expressed entirely through the retarded GF

$$I = \frac{e}{h}\int dE(f_L - f_R)\mathrm{Tr}[\Gamma_L(E)G_{LR}(E)\Gamma_R(E)G^{\dagger}_{RL}(E)], \tag{16}$$

where the integrand $\mathcal{T} = \mathrm{Tr}[\Gamma_L G^0_{LR}\Gamma_R G^{0\dagger}_{RL}]$ is the transmission probability. This formula is originally due to Caroli *et al.*,[7] who derived it in the absence of disorder. Later Meir and Wingreen showed that it is valid for any noninteracting electron system in an arbitrary external potential, including disorder.[8]

Our objective here is to calculate the current through a disordered medium. For any particular disorder configuration we can obtain the full G and F and calculate the current via Eqs. (4) or (16) which will give identical results. Since for any particular configuration the two expressions give the same result, the configurational average over all disorder configurations must be the same,

$$\begin{aligned}\langle I\rangle &\sim \int dE \sum_j \mathrm{Tr}[H_{i+1,j;i,j}\langle F_{i,j;i+1,j}\rangle - H_{i,j;i+1,j}\langle F_{i+1,j;i,j}\rangle] \\ &= \int dE(f_L - f_R)\mathrm{Tr}[\langle\Gamma_L G_{LR}\Gamma_R G^{\dagger}_{RL}\rangle],\end{aligned} \tag{17}$$

which means that we could either account for disorder through the configuration average of the Keldysh GF or through taking the product and subsequent configurational average of the retarded and advanced GFs. Both methods will give identical results regardless of how disorder is treated.

A. CPA: Standard formulation

First we will calculate the current using the standard CPA technique for G and Eq. (16) for the current. We consider the classical nonmagnetic binary alloy case when two atomic species A and B randomly occupy each lattice site with probabilities p and $1-p$ respectively. The method is easily generalizable for other types of disorder. The disordered medium can be described by $H = H'_0 + V$, where $H'_0 = H_0 + \Sigma_L + \Sigma_R$ is the periodic part of the Hamiltonian and V is the deviation from periodicity. Typically the periodic part is given by the average Hamiltonian $H_0 = \langle H\rangle$ which is the spirit of the virtual crystal approximation (VCA).[2] If we consider only on-site disorder $\sum_n V_n c^{\dagger}_n c_n$, the retarded GF of the disordered medium is given by the Dyson equation

$$G_{nm} = G^0_{nm} + \sum_l G^0_{nl}V_l G_{lm}. \tag{18}$$

The essence of the CPA is to replace the disordered medium with a periodic effective medium described by a yet unknown coherent potential $\bar{H} = H'_0 + \Sigma$, where Σ is on-site. The coherent potential Σ is a complex energy-dependent quantity which physically can be viewed as the self-energy due to disorder. Then the Dyson equation can be written through the GF of the effective medium $\bar{G} = (E - \bar{H})^{-1}$ as

$$G_{nm} = \bar{G}_{nm} + \sum_l \bar{G}_{nl}(V_l - \Sigma)G_{lm}, \tag{19}$$

where $\bar{G}_{nm} = G^0_{nm} + \sum_l G^0_{nl}\Sigma\bar{G}_{lm}$. Expanding the equation into infinite series we obtain[2]

$$G_{nm} = \bar{G}_{nm} + \sum_l \bar{G}_{nl}t_l\bar{G}_{lm} + \sum_{l,l'}\bar{G}_{nl}t_l\bar{G}_{ll'}t_{l'}\bar{G}_{l'm} + \cdots, \tag{20}$$

where t_l is the single-site scattering matrix

$$t_l = (V_l - \Sigma)[1 - \bar{G}_{ll}(V_l - \Sigma)]^{-1}, \tag{21}$$

and it is understood that in the expansion consecutive t's are on the different sites. We can define the full retarded scattering matrix

$$T_{ll'} = t_l\delta_{ll'} + t_l\bar{G}_{ll'}t_{l'} + \sum_k t_l\bar{G}_{lk}t_k\bar{G}_{kl'}t_{l'} + \cdots, \tag{22}$$

which encapsulates the information for all scattering events. With the help of the full t matrix we can rewrite Eq. (20) only as a function of the GF of the effective medium

$$G_{nm} = \bar{G}_{nm} + \sum_{ll'}\bar{G}_{nl}T_{ll'}\bar{G}_{l'm}. \tag{23}$$

In order to obtain the unknown coherent potential Σ we impose the condition that the scattering vanishes on average in the effective medium $\langle T\rangle = 0$. Satisfying this condition means that the average GF coincides with that of the effective medium $\langle G\rangle = \bar{G}$. Practically it is difficult to satisfy this condition for the full t matrix because it would mean setting $\langle t_{l_1}t_{l_2}..t_{l_m}\rangle = 0$ to arbitrary order. Within the single-site approximation to CPA, correlations of electron scattering between different sites are neglected which means that $T = T(\langle t\rangle)$ only. Thus, the requirement that the single-site t matrix vanishes on average,

$$\langle t_l\rangle = pt^A_l + (1-p)t^B_l = 0, \tag{24}$$

guarantees that $\langle T\rangle = 0$. In effect the single-site approximation means that scattering from a single site is included exactly but scattering from two or more sites is neglected. In diagrammatic terms this means that all crossing diagrams in the expansion for $\langle T\rangle$ are ignored and the only contribution comes from the ladder diagrams.[2,23]

The procedure for evaluating the coherent potential is as follows: We work in the two-probe setup in which the current flows along the z axis and system is periodic in the xy plane perpendicular to the current direction. Therefore, we use the mixed ($\mathbf{k}_{\parallel}$,z) representation and perform a Fourier transform in the plane. In this representation the GF of the effective medium is

$$\bar{G}(\mathbf{k}_{\parallel}) = [I - G^0(\mathbf{k}_{\parallel})\Sigma]^{-1}G^0(\mathbf{k}_{\parallel}), \tag{25}$$

where the coherent potential is independent of $\mathbf{k}_{\parallel}$ (however, it is layer dependent). Then we apply the requirement Eq. (24) that the average single-site t matrix is equal to zero for any

layer l in the scattering region

$$0 = pt_l^A + (1-p)t_l^B = p\left(V_l^A - \Sigma_l\right)\left[I - \bar{G}_{ll}\left(V_l^A - \Sigma_l\right)\right]^{-1} + (1-p)\left(V_l^B - \Sigma_l\right)\left[I - \bar{G}_{ll}\left(V_l^B - \Sigma_l\right)\right]^{-1}, \quad (26)$$

where $\bar{G}_{ll} = \frac{1}{V_{BZ}} \int d\mathbf{k}_\parallel \bar{G}_{ll}(\mathbf{k}_\parallel)$ is the inverse Fourier transform of the retarded GF on site l. Equations (25) and (26) are a nonlinear system of equations for $\bar{G}$ and Σ. We solve this system for all disordered layers simultaneously using nonlinear solvers such as Newton-Raphson. The coherent potential is in general complex and energy dependent through $\bar{G}$.

After obtaining the coherent potential (and respectively the GF of the effective medium) we can proceed to calculate the transmission. Let indices a (b) denote the first principal layer of the barrier to the left (right). The formula for the transmission probability in real space is

$$\mathcal{T} = \mathrm{Tr}[\Gamma_a(\rho_1,\rho_2)G_{ab}(\rho_2,\rho_3)\Gamma_b(\rho_3,\rho_4)G^\dagger_{ba}(\rho_4,\rho_1)], \quad (27)$$

where the trace is a shorthand for the sum over all repeating indices. Making use of the periodicity in the plane we Fourier transform as follows: $\bar{G}(\rho,\rho') = \frac{1}{N}\sum_{\mathbf{k}_\parallel} \bar{G}(\mathbf{k}_\parallel)\exp[i\mathbf{k}_\parallel(\rho - \rho')]$, and the same for Γ. Next we calculate the configuration average of the transmission probability

$$\langle\mathcal{T}\rangle = \mathrm{Tr}\langle\Gamma_L G \Gamma_R G^\dagger\rangle = \mathrm{Tr}[\Gamma_L \bar{G}\Gamma_R\bar{G}^\dagger] + \mathrm{Tr}[\Gamma_L\bar{G}\langle T\bar{G}\Gamma_R\bar{G}^\dagger T^\dagger\rangle\bar{G}^\dagger]. \quad (28)$$

Substituting in this expression the expansion of the scattering matrix Eq. (22) we obtain contributions to the transmission probability to infinite order

$$\langle\mathcal{T}(E)\rangle = \mathcal{T}^{(0)} + \mathcal{T}^{(1)} + \mathcal{T}^{(2)} + \cdots, \quad (29)$$

where all the terms contain only the retarded GF of the effective medium $\bar{G}$ and the single-site scattering matrix t_l, in addition to the escape rates Γ which do not depend on the disorder. The first term in the expansion is the so-called "bubble" diagram

$$\mathcal{T}^{(0)} = \mathrm{Tr}[\Gamma_a(\rho_1,\rho_2)\bar{G}_{ab}(\rho_2,\rho_3)\Gamma_b(\rho_3,\rho_4)\bar{G}^\dagger_{ba}(\rho_3,\rho_4)] = \int d\mathbf{k}_\parallel \mathrm{Tr}[\Gamma_a(\mathbf{k}_\parallel)\bar{G}_{ab}(\mathbf{k}_\parallel)\Gamma_b(\mathbf{k}_\parallel)\bar{G}^\dagger_{ba}(\mathbf{k}_\parallel)], \quad (30)$$

which does not depend on the disorder configuration and represents the transmission of the effective medium. Since the effective medium is periodic in the plane, $\mathbf{k}_\parallel$ is a good quantum number and the transmission is specular. Vertex corrections are the diagrams of higher order in t_l. The first-order correction is

$$\mathcal{T}^{(1)} = \mathrm{Tr}[\langle\Gamma_a(\rho_1,\rho_2)\bar{G}_{al}(\rho_2,\rho_3)t_l(\rho_3)\bar{G}_{lb}(\rho_3,\rho_4) \times \Gamma_b(\rho_4,\rho_5)\bar{G}^\dagger_{bl}(\rho_5,\rho_6)t_l^\dagger(\rho_6)\bar{G}^\dagger_{la}(\rho_6,\rho_1)\rangle] = \int d\mathbf{k}_{1\parallel}d\mathbf{k}_{2\parallel}\mathrm{Tr}[\Gamma_a(\mathbf{k}_{1\parallel})\bar{G}_{al}(\mathbf{k}_{1\parallel}) \times \langle t_l\bar{G}_{lb}(\mathbf{k}_{2\parallel})\Gamma_b(\mathbf{k}_{2\parallel})\bar{G}^\dagger_{bl}(\mathbf{k}_{2\parallel})t_l^\dagger\rangle\bar{G}^\dagger_{la}(\mathbf{k}_{1\parallel})], \quad (31)$$

which involves processes of electron transmission from a state with momentum $\mathbf{k}_{1\parallel}$ to a state with momentum $\mathbf{k}_{2\parallel}$; i.e., the transmission is diffusive. Similarly, expressions can be derived for the higher order diagrams and the expressions become increasingly involved.

B. CPA: Nonequilibrium formulation

Next we calculate the current applying the NE-CPA technique for F and then using Eq. (4) for the current. Within the NEGF formalism the full GF for a disordered system obeys a generalized Dyson equation[19]

$$\hat{G}_{nm} = \hat{G}^0_{nm} + \sum_l \hat{G}^0_{nl}\hat{V}_l\hat{G}_{lm}, \quad (32)$$

where $\hat{G} = \begin{pmatrix} G^c & G^< \\ G^> & \tilde{G}^c \end{pmatrix}$ and $\hat{V} = \begin{pmatrix} V^c & V^< \\ V^> & \tilde{V}^c \end{pmatrix}$. The various NEGFs are defined as usual:[19]

$$iG^c_{12} = \langle T\hat{\Psi}_1\hat{\Psi}_2^\dagger\rangle, \quad i\tilde{G}^c_{12} = \langle\tilde{T}\hat{\Psi}_1\hat{\Psi}_2^\dagger\rangle, \quad iG^<_{12} = -\langle\hat{\Psi}_2^\dagger\hat{\Psi}_1\rangle, \quad iG^>_{12} = \langle\hat{\Psi}_1\hat{\Psi}_2^\dagger\rangle, \quad (33)$$

where $1 = (t_1,\mathbf{r}_1)$ and $2 = (t_2,\mathbf{r}_2)$ and T and $\tilde{T}$ mean chronological and reverse-chronological ordering of the field operators. Since the perturbation comes from a single-particle time-independent operator V, one can obtain explicitly the general form using the Keldysh perturbation technique $\hat{V} = \begin{pmatrix} V & 0 \\ 0 & -V \end{pmatrix}$. Since the components of $\hat{G}$ are not linearly independent, we use the Keldysh linear transformation $\hat{G}' = R^{-1}\hat{G}R$, where $R = \frac{1}{2}\begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix}$, to obtain an equation for $\hat{G}' = \begin{pmatrix} 0 & G^\dagger \\ G & F \end{pmatrix}$ and $\hat{V}' = \begin{pmatrix} 0 & V \\ V & 0 \end{pmatrix}$.[19] Here G and $G^\dagger$ are the retarded and advanced GF and F is the Keldysh GF defined as follows:

$$G = G^c - G^< = -\tilde{G}^c + G^>, \quad F = G^c + \tilde{G}^c = G^> + G^<. \quad (34)$$

Since the equations for the retarded and the advanced GF are equivalent, the transformed generalized Dyson equation is equivalent to two independent equations, the Dyson equation for the retarded GF

$$G_{nm} = G^0_{nm} + \sum_l G^0_{nl}V_lG_{lm} \quad (35)$$

and the quantum kinetic equation for the Keldysh GF[19]

$$F_{nm} = F^0_{nm} + \sum_l F^0_{nl}V_l^\dagger G^\dagger_{lm} + \sum_l G^0_{nl}V_lF_{lm}. \quad (36)$$

At this point we carry out the CPA recipe for the NEGF. We start from the generalized Dyson equation for disordered system Eq. (32) where we replace the disordered medium out of thermodynamical equilibrium with a periodic effective medium described by a yet unknown generalized coherent potential $\hat{\Sigma} = \begin{pmatrix} \Sigma^c & \Sigma^< \\ \Sigma^> & \tilde{\Sigma}^c \end{pmatrix}$ as follows:

$$\hat{G}_{nm} = \hat{\bar{G}}_{nm} + \sum_l \hat{\bar{G}}_{nl}(\hat{V}_l - \hat{\Sigma})\hat{G}_{lm}. \quad (37)$$

After transformation $\hat{\Sigma}' = R^{-1}\hat{\Sigma}R = \begin{pmatrix} \Omega & \Sigma \\ \Sigma^\dagger & 0 \end{pmatrix}$. This generalized coherent potential consists of the standard coherent potential Σ and the nonequilibrium coherent potential Ω. Physically Ω can be viewed as the nonequilibrium part of the self-energy due to disorder. The two independent equations of the transformed generalized Dyson equations become Eq. (19)

for the retarded GF already considered and

$$F_{nm} = \bar{F}_{nm} - \sum_l \bar{G}_{nl}\Omega G^\dagger_{lm} + \sum_l \bar{F}_{nl}(V_l - \Sigma^\dagger)G^\dagger_{lm} + \sum_l \bar{G}_{nl}(V_l - \Sigma)F_{lm}, \quad (38)$$

where $\bar{F}_{nm} = F^0_{nm} + \sum_l (G^0_{nl}\Omega\bar{G}^\dagger_{lm} + F^0_{nl}\Sigma^\dagger\bar{G}^\dagger_{lm} + G^0_{nl}\Sigma\bar{F}_{lm})$. Next we rewrite Eq. (38) as the infinite expansion of contributions containing the retarded, advanced, and Keldysh GFs of the effective medium

$$\begin{aligned} F_{nm} = \bar{F}_{nm} &+ \sum_l (\bar{G}_{nl}\tau_l\bar{G}^\dagger_{lm} + \bar{F}_{nl}t^\dagger_l\bar{G}^\dagger_{lm} + \bar{G}_{nl}t_l\bar{F}_{lm}) \\ &+ \sum_{l,l'} (\bar{G}_{nl}t_l\bar{G}_{ll'}\tau_{l'}\bar{G}^\dagger_{l'm} + \bar{G}_{nl}\tau_l\bar{G}^\dagger_{ll'}t^\dagger_{l'}\bar{G}^\dagger_{l'm} \\ &+ \bar{G}_{nl}t_l\bar{F}_{ll'}t^\dagger_{l'}\bar{G}^\dagger_{l'm} + \bar{F}_{nl}t^\dagger_l\bar{G}^\dagger_{ll'}t^\dagger_{l'}\bar{G}^\dagger_{l'm} \\ &+ \bar{G}_{nl}t_l\bar{G}_{ll'}t_{l'}\bar{F}^\dagger_{l'm}) + \cdots, \end{aligned} \quad (39)$$

where by analogy with t_l we have defined a nonequlibrium single-site t matrix

$$\tau_l = t_l\bar{F}_{ll}t^\dagger_l - t_l(V_l - \Sigma)^{-1}\Omega(V_l - \Sigma^\dagger)^{-1}t^\dagger_l. \quad (40)$$

Note that in Eq. (39) consecutive t's and τ's are on the different sites. Next we introduce the Keldysh counterpart of the full scattering matrix

$$T^F_{ll'} = \tau_l\delta_{ll'} + t_l\bar{G}_{ll'}\tau_{l'} + \tau_l\bar{G}^\dagger_{ll'}t^\dagger_{l'} + t_l\bar{F}_{ll'}t^\dagger_{l'} + \cdots \quad (41)$$

and Eq. (39) can be written through the full t matrices and the GFs of the effective medium

$$F_{nm} = \bar{F}_{nm} + \sum_{ll'} \left(\bar{G}_{nl}T^F_{ll'}\bar{G}^\dagger_{l'm} + \bar{F}_{nl}T^\dagger_{ll'}\bar{G}^\dagger_{l'm} + \bar{G}_{nl}T_{ll'}\bar{F}_{l'm}\right). \quad (42)$$

Eventually we conclude that the NEGF can be written through the NEGF of the effective medium

$$\hat{G}_{nm} = \hat{\bar{G}}_{nm} + \sum_{ll'} \hat{\bar{G}}_{nl}\hat{T}_{ll'}\hat{\bar{G}}_{l'm}, \quad (43)$$

where the full nonequilibrium scattering matrix is defined as

$$\hat{T}_{ll'} = \hat{t}_l\delta_{ll'} + \hat{t}_l\hat{\bar{G}}_{ll'}\hat{t}_{l'} + \sum_k \hat{t}_l\hat{\bar{G}}_{lk}\hat{t}_k\hat{\bar{G}}_{kl'}\hat{t}_{l'} + \cdots, \quad (44)$$

and $\hat{T} = \left(\begin{smallmatrix} T^F & T \\ T^\dagger & 0 \end{smallmatrix}\right)$ and $\hat{t}_l = \left(\begin{smallmatrix} \tau_l & t_l \\ t^\dagger_l & 0 \end{smallmatrix}\right)$ are the full and the single-site t matrices of the effective medium.

Then following the prescription, we apply the requirement that scattering vanishes on average in the effective medium $\langle\hat{T}\rangle = 0$ which implies that both $\langle T\rangle = \langle T^F\rangle = 0$. Within the single-site approximation the NE-CPA condition becomes $\langle\hat{t}_l\rangle = 0$ which is equivalent to two independent conditions, Eq. (24) for t_l and the additional condition

$$\langle\tau_l\rangle = 0. \quad (45)$$

The procedure for solving for the nonequilibrium coherent potential is as follows: (i) The CPA condition Eq. (26) combined with Eq. (25) allows us to evaluate Σ identically to the standard CPA. (ii) After Σ is obtained, we write down the quantum kinetic equation in the matrix notation for the Keldysh GF of the effective medium

$$\begin{aligned} \bar{F}(\mathbf{k}_\parallel) = F^0(\mathbf{k}_\parallel) &+ G^0(\mathbf{k}_\parallel)\Omega\bar{G}^\dagger(\mathbf{k}_\parallel) \\ &+ F^0(\mathbf{k}_\parallel)\Sigma^\dagger\bar{G}^\dagger(\mathbf{k}_\parallel) + G^0(\mathbf{k}_\parallel)\Sigma\bar{F}(\mathbf{k}_\parallel), \end{aligned} \quad (46)$$

where the nonequilibrium coherent potential Ω is independent on $\mathbf{k}_\parallel$. Now we can combine the terms involving $\bar{F}$ to obtain

$$\begin{aligned} \bar{F}(\mathbf{k}_\parallel) = \bar{G}(\mathbf{k}_\parallel)&\Omega\bar{G}^\dagger(\mathbf{k}_\parallel) \\ &+ [I - G^0(\mathbf{k}_\parallel)\Sigma]^{-1}F^0(\mathbf{k}_\parallel)[I + \Sigma^\dagger\bar{G}^\dagger(\mathbf{k}_\parallel)]. \end{aligned} \quad (47)$$

The second equation is obtained from Eq. (45) that the average of the nonequilibrium single-site scattering matrix $\langle\tau_l\rangle$ is equal to zero for any layer l in the scattering region

$$\begin{aligned} 0 &= p\tau^A_l + (1-p)\tau^B_l \\ &= p\left[t^A_l\bar{F}_{ll}t^{A\dagger}_l - t^A_l\left(V^A_l - \Sigma\right)^{-1}\Omega\left(V^A_l - \Sigma^\dagger\right)^{-1}t^{A\dagger}_l\right] \\ &\quad + (1-p)\left[t^B_l\bar{F}_{ll}t^{B\dagger}_l - t^B_l\left(V^B_l - \Sigma\right)^{-1}\Omega\left(V^B_l - \Sigma^\dagger\right)^{-1}t^{B\dagger}_l\right], \end{aligned} \quad (48)$$

where $\bar{F}_{ll} = \frac{1}{V_{BZ}}\int d\mathbf{k}_\parallel \bar{F}_{ll}(\mathbf{k}_\parallel)$ is the inverse Fourier transform of the Keldysh GF at layer l. We solve the *linear* system of Eqs. (47) and (48) to determine $\bar{F}$ and Ω. Practically, it is much easier to solve linear Eqs. (47) and (48) for the NE-CPA than the nonlinear system of Eqs. (25) and (26) for the standard CPA. Also problems with convergence to the wrong solutions do not arise as could happen in the nonlinear solver. Finally, the average NEGF is equal to the NEGF of the effective medium $\langle F\rangle = \bar{F}$.

Using Eq. (38) and Eq. (4), the average transmission is simply related to the Keldysh GF of the effective medium

$$\langle\mathcal{T}\rangle = \sum_j \mathrm{Tr}[H_{i+1,j;i,j}\bar{F}_{i,j;i+1,j} - H_{i,j;i+1,j}\bar{F}_{i+1,j;i,j}]. \quad (49)$$

Therefore, it follows that the nonequilibrium transmission probability given by Eq. (49) is equivalent to that given by Eq. (28) which contains VCs to infinite order.

III. EXAMPLE: TUNNEL JUNCTION WITH DISORDERED BARRIER

In order to demonstrate the method we consider a simple example consisting of a tunnel junction with a plane of impurities within the barrier (Fig. 1). We use a single-band tight-binding model characterized with on-site energy ε and hopping integral t. Despite its simplicity this model has been shown to be capable of predicting the correct properties of several phenomena including spin transfer torque,[24–28] interlayer exchange coupling,[29] and resistive switching phenomena.[30] Its predictions compare well to the results of first-principles calculations.[31] The two electrodes are assumed to be locally in equilibrium at two different chemical potentials μ_L and μ_R under finite bias $\mu_L - \mu_R = eV$, where V is the applied voltage. Under this assumption electrons will flow from the electrode with higher chemical potential to that with a lower. For simplicity's sake we assume that the voltage drop occurs linearly across the barrier, which is accounted for in the Hamiltonian via an external potential term $\sum_i eV_i c^\dagger_i c_i$. The electronic temperature is taken into account via the Fermi-Dirac

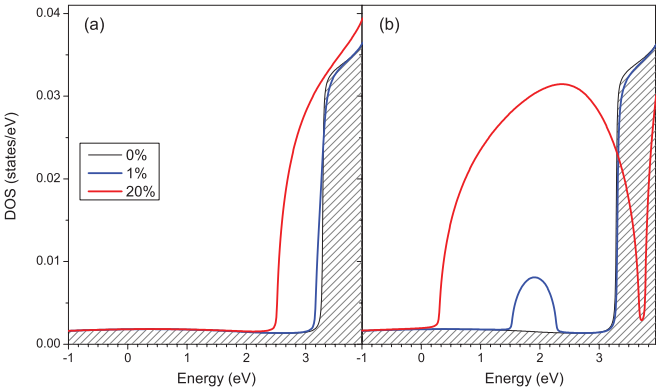

FIG. 2. (Color online) Density of states in the barrier for weak (a) and strong (b) scattering. Both low and high impurity concentration are shown.

distribution functions $f_{L/R} = [1 + \exp(E - \mu_{L(R)})/kT]^{-1}$. The atoms are at zero temperature and stay fixed at their equilibrium positions. In the electrodes the on-site energy $\varepsilon_0 = 0$ eV can be considered as a reference. Then in the barrier we choose $\varepsilon = 9$ eV. For the impurity we consider two cases: (i) weak scattering ($\varepsilon' = 6$ eV) and (ii) strong scattering ($\varepsilon' = 3$ eV). Hopping is the same between all sites $t = 1$ eV. The simplest structure has five layers in the barrier where the impurity layer is in the middle. Two impurity concentrations are considered low: ($p = 1\%$) and high ($p = 20\%$). We calculate the I-V curves when we apply voltage of up to 1.0 V symmetrically. The system is considered to be at low temperature.

The density of states (DOS) of the electrodes consists of a metallic band centered at 0 with a dispersion of ± 6 eV. At finite bias the left band is shifted down by $-\Delta V/2$ and the right up by $\Delta V/2$. The DOS in the barrier (Fig. 2) shows the onset of the conduction band at around 3 eV. Then the impurity bands appear around 6 and 3 eV for weak and strong scattering, respectively. For weak scattering it overlaps with the conduction band and the net effect is to modify the conduction band minimum. For strong scattering it appears as a separate band. The impurity band is small and narrow at low concentrations and becomes larger and wider at high concentrations. The tail of the impurity band will influence the transmission and we expect larger effect when the impurity band is closer to the Fermi level and resonance scattering when the impurity band falls in the bias window.

We calculate the current using (i) the standard CPA approach with vertex corrections to infinite order and (ii) nonequilibrium CPA. For our toy model the various quantities are scalars which commute with each other. Due to this simplification it is possible to derive a formula for the vertex corrections to any finite order and also sum all the corrections exactly to infinite order. The first-order correction becomes

$$\mathcal{T}^{(1)} = \langle |t_l|^2 \rangle \int d\mathbf{k}_\parallel \bar{G}^\dagger_{la}(\mathbf{k}_\parallel)\Gamma_a(\mathbf{k}_\parallel)\bar{G}_{al}(\mathbf{k}_\parallel) \times \int d\mathbf{k}_\parallel \bar{G}_{lb}(\mathbf{k}_\parallel)\Gamma_b(\mathbf{k}_\parallel)\bar{G}^\dagger_{bl}(\mathbf{k}_\parallel). \quad (50)$$

If we continue to write explicitly the higher order VCs, a patten emerges (after some lengthy algebra) which allows us to calculate the higher order corrections recursively as follows:

$$\mathcal{T}^{(n)} = \mathcal{T}^{(n-1)} Q_l, \quad Q_l = \langle |t_l|^2 \rangle \left[\int d\mathbf{k}_\parallel |\bar{G}_{ll}(\mathbf{k}_\parallel)|^2 - \left| \int d\mathbf{k}_\parallel \bar{G}_{ll}(\mathbf{k}_\parallel) \right|^2 \right], \quad (51)$$

which implies that the vertex corrections to infinite order can be summed exactly as a geometric progression

$$\langle \mathcal{T} \rangle = \mathcal{T}^0 + \mathcal{T}^{(1)}/(1 - Q_l) \quad (52)$$

and if the term $|Q_l| < 1$ the sum converges.

An additional advantage of the toy model of a single-band model and one impurity layer is that we can obtain an analytical expression for Ω. Following the NE-CPA method we solve Eqs. (47) and (48) analytically using the linear dependence of $\bar{F}$ on Ω

$$\bar{F}_{ll} = \alpha + \beta\Omega, \quad \Omega = \alpha\big(V_l^A - \Sigma\big)\big(V_l^B - \Sigma^\dagger\big)\big[\big(V_l^A - \Sigma\big)\bar{G}_{ll} + \bar{G}^\dagger_{ll}\big(V_l^B - \Sigma^\dagger\big) - \beta\big(V_l^A - \Sigma\big)\big(V_l^B - \Sigma^\dagger\big) - 1\big]^{-1}, \quad (53)$$

where the coefficients are

$$\alpha = \int \frac{d\mathbf{k}_\parallel}{(2\pi)^2} F^0_{ll}(\mathbf{k}_\parallel)\{1 + 2\mathrm{Re}[\Sigma\bar{G}_{ll}(\mathbf{k}_\parallel)] + |\Sigma\bar{G}_{ll}(\mathbf{k}_\parallel)|^2\}, \quad \beta = \int \frac{d\mathbf{k}_\parallel}{(2\pi)^2} G^0_{ll}(\mathbf{k}_\parallel)\bar{G}^\dagger_{ll}(\mathbf{k}_\parallel)[1 + \Sigma\bar{G}_{ll}(\mathbf{k}_\parallel)]. \quad (54)$$

The I-V curves calculated using both approaches are shown in Fig. 3 for the strong-scattering case with high impurity concentration. In the first case we calculate the current due to the bubble term plus vertex corrections to infinite order. The contribution from different orders of the VCs is also plotted for comparison. From the results several observations can be made: (i) When the impurity band is far from the bias window CPA alone gives a reasonable result and the VCs converge very quickly. (ii) However, if the the impurity band is in the bias window CPA captures only a very small part of the part of the contribution to the current. Also the higher order vertex

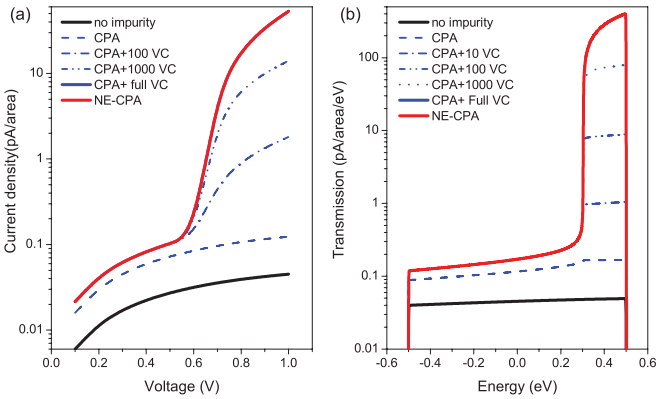

FIG. 3. (Color online) Current density as a function of applied bias (a) and transmission as a function of energy (b) for strong scattering and high impurity concentration. The result for CPA, CPA with VCs to finite order, CPA with VCs to infinite order, and NE-CPA are plotted. The line of the CPA with full VCs coincides with the NE-CPA line.

corrections are by no means small, which means that the expansion is not perturbative. In particular for strong scattering summation to infinite order is necessary to correctly describe the transmission. This confirms the importance of the vertex corrections.[9,10] (iii) NE-CPA always gives the same result as the standard CPA with VCs to infinite order.

IV. CONCLUSIONS

In summary, we propose a nonequilibrium formulation of CPA for electron transport. This approach makes use of the fact that the nonequilibrium current is linear with respect to the NEGF and therefore the average current is simply expressed through the GF of the effective medium. This removes the need to calculate vertex corrections to infinite order. The single-step NE-CPA formalism is equivalent to CPA plus vertex corrections to infinite order. This is demonstrated in a model calculation of a tunnel barrier with an impurity layer for a range of concentrations and scattering strengths. The NE-CPA is formulated in matrix form and therefore it is easily extendable to general LCAO Hamiltonians, including magnetic. Indeed the approach would show its true worth when implemented within the framework of more realistic electronic structure calculations.

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[1] R. Elliott, J. A. Krumhansl, and P. L. Leath, Rev. Mod. Phys. **46**, 465 (1974).
[2] E. N. Economou, *Green's Functions in Quantum Physics* (Springer, 2010).
[3] P. Soven, Phys. Rev. **156**, 809 (1967).
[4] D. W. Taylor, Phys. Rev. **156**, 1017 (1967).
[5] B. Velicky, S. Kirkpatrick, and H. Ehrenreich, Phys. Rev. **175**, 747 (1968).
[6] B. Velicky, Phys. Rev. **184**, 614 (1969).
[7] C. Caroli, R. Combescot, P. Nozieres, and D. Saint-James, J. Phys. C **4**, 916 (1971).
[8] I. Meir and N. Wingreen, Phys. Rev. Lett. **68**, 2512 (1992).
[9] V. Drchal, J. Kudrnovský, P. Bruno, P. H. Dederichs, I. Turek, and P. Weinberger, Phys. Rev. B **65**, 214414 (2002).
[10] J. Velev and W. H. Butler, J. Appl. Phys. **97**, 10C517 (2005).
[11] S. V. Faleev, F. Leonard, D. A. Stewart, and M. van Schilfgaarde, Phys. Rev. B **71**, 195422 (2005).
[12] H. Itoh, A. Shibata, T. Kumazaki, J. Inoue, and S. Maekawa, J. Phys. Soc. Jpn. **68**, 1632 (1999).
[13] M. Ye. Zhuravlev, A. V. Vedyayev, K. D. Belashchenko, and E. Y. Tsymbal, Phys. Rev. B **85**, 115134 (2012).
[14] G. M. Stocks and H. Winter, Z. Phys. B **46**, 95 (1982).
[15] W. H. Butler, Phys. Rev. B **31**, 3260 (1985).
[16] J. Kudrnovsky, V. Drchal, and J. Masek, Phys. Rev. B **35**, 2487 (1987).
[17] J. Kudrnovsky and V. Drchal, Phys. Rev. B **41**, 7515 (1990).
[18] Y. Ke, K. Xia, and H. Guo, Phys. Rev. Lett. **100**, 166805 (2008).
[19] E. M. Lifshitz and L. P. Pitaevskii, *Physical Kinetics*, Course of Theoretical Physics, Vol. 10 (Pergamon, Oxford, 1981).
[20] J. Velev and W. Butler, J. Phys.: Condens. Matter **16**, R637 (2004).
[21] S. Datta, *Electronic Transport in Mesoscopic Systems* (Cambridge University Press, 1995).
[22] A. Kalitsov, M. Chshiev, and J. Velev (unpublished).
[23] K. M. Schep and G. E. W. Bauer, Phys. Rev. B **56**, 15860 (1997).
[24] I. Theodonis, N. Kioussis, A. Kalitsov, M. Chshiev, and W. H. Butler, Phys. Rev. Lett. **97**, 237205 (2006).
[25] M. Chshiev, I. Theodonis, A. Kalitsov, N. Kioussis, and W. H. Butler, IEEE Trans. Magn. **44**, 2453 (2008).
[26] A. Kalitsov, M. Chshiev, I. Theodonis, N. Kioussis, and W. H. Butler, Phys. Rev. B **79**, 174416 (2009).
[27] H. Kubota, A. Fukushima, K. Yakushiji, T. Nagahama, S. Yuasa, K. Ando, H. Maehara, Y. Nagamine, K. Tsunekawa, D. Djayaprawira, N. Watanabe, and Y. Suzuki, Nat. Phys. **4**, 37 (2008).
[28] J. Sankey, Y-T. Cui, J. Sun, J. Slonczewski, R. Buhrman, and D. Ralph, Nat. Phys. **4**, 67 (2008).
[29] H. X. Yang, M. Chshiev, A. Kalitsov, A. Schuhl, and W. H. Butler, Appl. Phys. Lett. **96**, 262509 (2010).
[30] A. Kalitsov, A. M. Sahadevan, S. N. Jammalamadaka, G. Kalon, C. Bhatia, G. Xiong, and H. Yang, AIP Adv. **1**, 042158 (2011).
[31] C. Heiliger and M. D. Stiles, Phys. Rev. Lett. **100**, 186805 (2008).