Scattering matrix theory for nonlinear transport

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We report a scattering matrix theory for dynamic and nonlinear transport in coherent mesoscopic conductors. In general this theory allows predictions of low-frequency linear dynamic conductance, as well as weakly nonlinear dc conductance. It satisfies the conditions of gauge invariance and electric current conservation, and can be put into a form suitable for numerical computation. Using this theory we examine the third-order weakly nonlinear dc conductance of a tunneling diode. [S0163-1829(98)03916-2]

I. INTRODUCTION

Quantum transport under a time-dependent field in coherent mesoscopic systems is the subject of many recent studies.[1–7] Another problem of interest is the nonlinear conductance of such a system, whether under a time-dependent field or not.[8] A difficult theoretical issue is the prediction, for a general mesoscopic conductor, for the transport coefficients as a function of the ac field frequency and the bias voltage. Once these parameters are known, one can predict useful information such as the nonlinear current-voltage characteristics in the dc case, the emittance in the linear frequency, linear voltage ac case, and further nonlinear dynamic conductance. Indeed, it is now possible to experimentally measure the nonlinear ac transport properties such as the second harmonic generation, as has been demonstrated by several laboratories.[9–11]

When a conductor is subjected to time-varying external fields such as an ac bias voltage, the total electric current flowing through the conductor consists of the usual particle current plus the displacement current. The presence of the latter is crucial, such that the total electric current is conserved. Hence, for a theory to deal with ac transport, in principle one should include the displacement current into the consideration. Because a displacement current originates from induction, and the necessary condition for electric induction is the electron-electron (e-e) interaction, one thus concludes that an important ingredient for ac transport theory should be the consideration of e-e interactions. These issues have been emphasized by Büttiker and Christen.[12] On the other hand, for dc transport under *nonlinear* conditions, a necessary requirement is the gauge invariance:[8] the physics should not change when electrostatic potential everywhere is changed by the same constant amount. Gauge invariance puts severe conditions on the form of the nonlinear transport coefficients. From these physical arguments, it is clear that ac as well dc nonlinear transport contains ingredients that were not needed when dealing with the familiar dc linear transport.[12]

The problems of current conservation and gauge invariance have been recognized in the literature. For conductors that maintain quantum coherence, Büttiker and his coworkers have developed an approach[2] based on the single electron scattering matrix theory to deal with the linear ac dynamic conductance as well as the second-order nonlinear conductance coefficients. The original scattering matrix theory was invented to investigate dc linear transport coefficients, as is represented by the Landauer-Büttiker formulation.[13] Such a theory calculates particle current from the scattering matrix, thus a direct application to the ac situation would violate current conservation.[2,12] To solve this problem, the scattering matrix theory for ac transport consists of two steps.[2,12] First, it calculates the particle current and finds that this current is not conserved. Second, it considers the e-e interaction that alters the scattering potential landscape, and this effect generates an internal response that cancels exactly the nonconserved part of the particle current thereby restoring the current conservation. For the dc second-order nonlinear conductance coefficient, similar considerations led to the desired gauge invariance.

In a recent work, the authors have developed a microscopic and general theoretical formalism for electric response that is appropriate for both dc and ac weakly nonlinear quantum transport.[14] That formalism was based on the response theory and it formalized the connection of the response theory to the scattering matrix theory at the weakly nonlinear level. One of the useful conceptual advances of the general formalism[14] was the introduction of a frequency-dependent characteristic potential at the nonlinear level. The characteristic potential describes the changes of scattering potential landscape of a mesoscopic conductor when the electrochemical potential of an electron reservoir is perturbed externally.[2] It is the nonlinear order characteristic potential that allowed us to analyze the weakly nonlinear ac response,[14] as well as the nonlinear dc conductance, order by order in the bias voltage. In contrast, so far the scattering matrix theory can be applied up to the second-order nonlinearity and linear order ac.

Using the concept of nonlinear characteristic potential developed in the response theory,[14] we have found that the scattering matrix theory can actually be further developed to apply to higher-order nonlinear dc situations. In addition, recognizing that an ac transport problem requires the self-consistent solution of the Schrödinger equation coupled with Maxwell equations, we have found a way to derive both the external and internal responses in *equal footing* within the scattering matrix approach. It is the purpose of this paper to report these results. In particular, we shall start from the scattering matrix theory and formulate an approach that is appropriate for analyzing linear order dynamic conductance and the weakly nonlinear dc conductance beyond the second order. We emphasize the properties of electric current conservation and gauge invariance, and these properties are maintained by considering electron-electron interactions. The approach developed here is particularly useful for nonlinear dc conductance calculations, and we shall analyze the third-order weakly nonlinear transport coefficient for a double-barrier tunneling diode. Since the approach presented here can be cast into a form that allows numerical computation, many further applications of it to complicated device structures can be envisioned.

The rest of the article is organized as follows. In the next section we present the development of the formalism. Section III presents two applications of this formalism: the linear ac dynamic conductance and the third-order nonlinear conductance. Finally, a short conclusion is included in Sec. IV.

II. THEORETICAL FORMALISM

In this section we briefly go through the formal development of our scattering matrix theory and concentrate more on the conceptually important physical quantities that will be needed.

We start by writing the Hamiltonian of the system in the presence of an external time-dependent field as

$$H=\sum_{\alpha m}\;(\overline{E}_{\alpha m}+eV_\alpha\;\cos\;\omega t)a^\dagger_{\alpha m}(\overline{E}_{\alpha m},t)a_{\alpha m}(\overline{E}_{\alpha m},t), \tag{1}$$

where $a^\dagger_{\alpha m}$ is the creation operator for a carrier in the incoming channel m in probe α, $eV_\alpha \cos\omega t$ is the shift of the electrochemical potential μ_α away from the equilibrium state associated with μ^{eq}, i.e., $eV_\alpha \cos\omega t=\mu_\alpha-\mu^{eq}$. The energy $\overline{E}_{\alpha m}$ is a functional of the internal electrical potential landscape $U(\mathbf{r},\{V_\alpha\})$ that depends on V_α in the low-frequency regime. Potential U includes the internal response to the external perturbation and it generates such effect as the displacement current. In general U is also an explicit function of time (or of the ac frequency ω) as discussed in Ref. 14, but in this work we shall only be concerned with the dynamic conductance to first power of ω, and for this case U is static. Note that we have explicitly included U into the Hamiltonian, which helps in dealing with both external and internal responses in equal footing. The self-consistent nature of this Hamiltonian is clear: U must be determined, in general, from the Maxwell equations where the charge density is obtained from solving the quantum-mechanical problem of the Hamiltonian.

Next let us consider the series expansion of the energy in terms of the potential landscape U,

$$\overline{E}_{\alpha m}+eV_\alpha\cos(\omega t)=E_{\alpha m}+e\hat{O}^{(1)}_\alpha\cos(\omega t)+e^2\hat{O}^{(2)}_\alpha\times[\cos(\omega t)]^2+\cdots, \tag{2}$$

where the operators $\hat{O}^{(i)}_\alpha$ are a spatial integration of the ith order *characteristic potential* (see below) folded with the ith order functional derivative of $E_{\alpha m}$ with respect to the potential landscape $U(\mathbf{r})$. For instance, the linear order operator, which is linear in voltage V_β, is given by

$$\hat{O}^{(1)}_\alpha\equiv\sum_\beta\;(\delta_{\alpha\beta}+\partial_{V_\beta}E_{\alpha m})V_\beta, \tag{3}$$

where

$$\partial_{V_\beta}E\;\equiv\;\int d^3\mathbf{r}\;\;u_\beta(\mathbf{r})\frac{\delta E}{\delta eU(\mathbf{r})}$$

with $u_\beta(\mathbf{r})\equiv\partial U(\mathbf{r})/\partial V_\beta$ the linear order characteristic potential.[2] The expressions for higher-order operators $\hat{O}^{(i)}_\alpha$ are more difficult to write down in a general form, but they are proportional to the ith power of the bias. In addition, they can be easily determined after we formally obtain the transmission function and then apply the current conservation and gauge invariance to the results. Using Eq. (2), the Hamiltonian now reads

$$H=\sum_{\alpha m}\left[E_{\alpha m}+\sum_i\;\hat{O}^{(i)}_\alpha(\cos\omega t)^i\right]a^\dagger_{\alpha m}(\overline{E}_{\alpha m},t)a_{\alpha m}(\overline{E}_{\alpha m},t). \tag{4}$$

The operators $a_{\alpha m}(\overline{E}_{\alpha m},t)$ satisfy the equation of motion

$$\dot{a}_{\alpha m}(\overline{E}_{\alpha m},t)=\frac{1}{i\hbar}[a_{\alpha m}(\overline{E}_{\alpha m},t),H], \tag{5}$$

which can be integrated because the time dependence of H is simple. For instance, to linear order in voltage, we only need to use $\hat{O}^{(1)}$ in the Hamiltonian and the result is

$$a_{\alpha,m}(\overline{E}_{\alpha m},t)=a_{\alpha m}(\overline{E}_{\alpha m})\times\exp\left(-\frac{i}{\hbar}\left[E_{\alpha m}t+\frac{e\hat{O}^{(1)}_\alpha}{\omega}\sin\;\omega t\right]\right).$$

Its Fourier transform is given by

$$\begin{aligned}\tilde{\mathbf{a}}_\alpha(E) &= \int dt\,\mathbf{a}_\alpha(\overline{E_\alpha},t)e^{iEt/\hbar}\\ &= \mathbf{a}_\alpha(E) - \frac{e}{2\hbar\omega}\hat{O}^{(1)}_\alpha[\mathbf{a}_\alpha(E+\hbar\omega)-\mathbf{a}_\alpha(E-\hbar\omega)]\\ &\quad + \frac{e^2}{8\hbar^2\omega^2}(\hat{O}^{(1)}_\alpha)^2[\mathbf{a}_\alpha(E+2\hbar\omega)\\ &\quad -2\mathbf{a}_\alpha(E)+\mathbf{a}_\alpha(E-2\hbar\omega)]+\cdots\\ &= \sum_n \frac{1}{n!}\left(\frac{-e\hat{O}^{(1)}_\alpha}{2\hbar\omega}\right)^n (e^{\hbar\omega\partial_E}-e^{-\hbar\omega\partial_E})^n\mathbf{a}_\alpha(E), \end{aligned} \tag{6}$$

where we have suppressed the index m and $\mathbf{a}_\alpha$ is in a vector form of the operators $a_{\alpha m}$. In Eq. (6) the physics is transparent: $\mathbf{a}_\alpha(E\pm\hbar\omega)$ is just the one-photon sideband and $\mathbf{a}_\alpha(E\pm2\hbar\omega)$ corresponds to the second harmonic generation. More tedious expressions can be obtained if higher-order operators $\hat{O}^{(i)}$ are included in the Hamiltonian.

To calculate the total electrical current, we shall apply the formula derived in Ref. 15, which is exact up to linear order of ω and for larger frequency it is an approximation to a space-dependent expression of the current operator

$$\begin{aligned} I_\alpha(t) &= \frac{e}{h}\int dE dE'[\tilde{\mathbf{a}}^\dagger_\alpha(E)\tilde{\mathbf{a}}_\alpha(E')-\tilde{\mathbf{b}}^\dagger_\alpha(E)\tilde{\mathbf{b}}_\alpha(E')]\\ &\quad \times \exp[i(E-E')t/\hbar], \end{aligned} \tag{7}$$

where $\tilde{\mathbf{b}}_\alpha(E)$ is the operator that annihilates a carrier in the outgoing channel in probe α. The annihilation operator in the outgoing channel $\tilde{\mathbf{b}}_\alpha$ is related to the annihilation operator in the incoming channel $\tilde{\mathbf{a}}_\alpha$ via the scattering matrix $\mathbf{s}_{\alpha\beta}$: $\tilde{\mathbf{b}}_\alpha = \Sigma_\beta \mathbf{s}_{\alpha\beta}\tilde{\mathbf{a}}_\beta$ where $\mathbf{s}_{\alpha\beta}$ is a function of energy E and a functional of the electric potential $U(\mathbf{r},\{V_\alpha\})$. Finally, we comment that in evaluating Eq. (7) we need to take a quantum statistical average of $\langle \mathbf{a}^\dagger_\alpha(E)\mathbf{a}_\beta(E')\rangle = \delta_{\alpha\beta}\delta(E-E')f_\alpha(E)$ where $f_\alpha(E)$ is the Fermi function of reservoir α. Because of the limitations of Eq. (7), our theory will be exact for transport coefficients linear in ω for ac situations. However this is not a severe limitation for practical calculations.[15]

One of the most important quantities of this theory is the determination of characteristic potential that arrives naturally. As discussed above, this quantity determines the operators $\hat{O}^{(i)}$. Since the scattering matrix theory used here is exact to the linear power of ω, which is the order we shall work on, we only need to consider ω-independent characteristic potentials. On the other hand, as we are interested in the weakly nonlinear coefficients, it is crucial to consider higher-order characteristic potentials:[14] $u_{\delta\gamma}(\mathbf{r}) \equiv \partial^2 U(\mathbf{r})/\partial V_\delta \partial V_\gamma$, $u_{\beta\delta\gamma}(\mathbf{r})$, etc. For any physical quantity beyond the terms linear in ω or second order in voltage, including the second-harmonic generation term (the term of ωV^2), these higher-order characteristic potentials are necessary.

We now discuss the solution of higher-order characteristic potentials by explicitly carrying out the calculation of $u_{\delta\gamma}$. In the weakly nonlinear regime, the variation of the electric potential can be expanded in terms of the variation of the electrochemical potential $d\mu$,

$$edU(\mathbf{r}) = \sum_\beta u_\beta(\mathbf{r})d\mu_\beta + \frac{1}{2}\sum_{\beta\gamma} u_{\beta\gamma}(\mathbf{r})d\mu_\beta d\mu_\gamma + \ldots, \tag{8}$$

where u_β is the characteristic potential, $u_{\beta\gamma}$ (which is symmetric in β and γ) is the second-order characteristic potential tensor, and $(\cdots)$ are higher-order terms written in a similar fashion. Because we are only interested in ac transport to the first power of frequency ω, the electrodynamics is solved by the Poisson equation

$$-\nabla^2 dU(\mathbf{r}) = 4\pi e^2 dn(\mathbf{r}) = 4\pi e^2\sum_\alpha dn_\alpha(\mathbf{r}), \tag{9}$$

where dn_α is the variation of the charge density at contact α due to a change in electrochemical potential at that contact. There are two contributions to the charge density at contact α: the injected charge density due to the variation of the chemical potential at contact α, and the induced charge density $dn_{ind,\alpha}$ due to the electrostatic potential. Hence,

$$dn_\alpha = \frac{dn_\alpha}{dE}d\mu_\alpha + \frac{1}{2}\frac{d^2n_\alpha}{dE^2}d\mu_\alpha^2 + \cdots + dn_{ind,\alpha}, \tag{10}$$

where dn_α/dE is the injectivity, which is the local density of state at contact α, and d^2n_α/dE^2 is the energy derivative of the injectivity. The induced charge density involves the Lindhard function. Using the Thomas-Fermi approximation, Eq. (10) takes a compact and simple form,

$$dn_\alpha = \frac{dn_\alpha}{dE}(d\mu_\alpha - edU) + \frac{1}{2}\frac{d^2n_\alpha}{dE^2}(d\mu_\alpha - edU)^2 + \cdots. \tag{11}$$

From Eqs. (8), (9), and (11), we obtain the equation satisfied by the second-order characteristic potential tensor

$$\begin{aligned} -\nabla^2 u_{\beta\gamma} + 4\pi e^2\frac{dn}{dE}u_{\beta\gamma} &= 4\pi e^2\left(\frac{d^2n_\beta}{dE^2}\delta_{\beta\gamma} - \frac{d^2n_\gamma}{dE^2}u_\beta \right.\\ &\quad \left. - \frac{d^2n_\beta}{dE^2}u_\gamma + \frac{d^2n}{dE^2}u_\beta u_\gamma\right). \end{aligned} \tag{12}$$

Since all the quantities involved in this equation are known from the linear order calculation, $u_{\beta\gamma}$ can thus be determined. Similarly, order by order we can determine higher-order characteristic potentials from results obtained at lower orders. For instance, the equation satisfied by $u_{\beta\gamma\delta}$ is found to be

$$\begin{aligned} &-\nabla^2 u_{\beta\gamma\delta} + 4\pi e^2\frac{dn}{dE}u_{\beta\gamma\delta}\\ &= 4\pi e^2\left(\frac{d^3n_\beta}{dE^3}\delta_{\beta\gamma}\delta_{\beta\delta} - \frac{d^3n}{dE^3}u_\beta u_\gamma u_\delta + \left\{\frac{d^3n_\beta}{dE^3}u_\gamma u_\delta \right.\right.\\ &\quad \left.\left. - \frac{d^3n_\beta}{dE^3}\delta_{\beta\gamma}u_\delta + \frac{d^3n}{dE^3}u_{\beta\gamma}u_\delta - \frac{d^3n_\beta}{dE^3}u_{\gamma\delta}\right\}_c\right), \end{aligned} \tag{13}$$

where the curly bracket $\{\ldots\}_c$ stands for the cyclic permutation of indices β, γ, and δ. Note that if better models are needed to deal with the screening effect, the term with dn/dE on the left-hand side of Eqs. (12) and (13) and higher-order equations is replaced by an integration over the appropriate Lindhard function folded with the characteristic potential.[14]

From Eq. (8) we can derive several important sum rules on the characteristic potential tensor. If all the changes in the electrochemical potentials are the same, i.e., $d\mu_\beta = d\mu_\gamma = d\mu$, this corresponds to an overall shift of the electrostatic potential $edU = d\mu$. From this we have $\Sigma_\beta u_\beta = 1$, $\Sigma_{\beta\gamma} u_{\beta\gamma} = 0$. Due to gauge invariance, Eq. (8) remains the same if dU, $d\mu_\beta$, and $d\mu_\gamma$ are all shifted by the same amount. This leads to $\Sigma_\beta u_{\beta\gamma} = \Sigma_\gamma u_{\beta\gamma} = 0$. Using Eq. (12), we can confirm that these relations are indeed satisfied. Similar sum rules can be derived for higher-order characteristic potentials.

Let's summarize the scattering matrix theoretical procedure. With the characteristic potential tensor calculated, we explicitly derive the Hamiltonian in a series from Eq. (4). The Hamiltonian determines the creation and annihilation operators via equation of motion Eq. (5) and the scattering matrix $\mathbf{s}_{\alpha\beta}$. Finally, using Eq. (7), we compute the electric current as a function of voltage.

III. APPLICATIONS

In the following we apply the scattering matrix formalism developed in the last section to two examples: the linear order emittance and the third-order dc nonlinear conductance. The first example has been examined by Büttiker and co-workers,[2] and our result is in exact agreement with theirs. The second example has not been studied and we shall provide further numerical results for a resonant tunneling diode.

A. Linear dynamic conductance

The linear dynamic conductance (called emittance) is the transmission function of the terms proportional to V_β and ωV_β in the electric current. From Eq. (7) we expand everything in these variables and obtain

$$I_\alpha(\omega') = \sum_\beta \hat{O}_\beta^{(1)} \int dE(-\partial_E f)\left\{ \frac{e^2}{h} A_{\beta\beta}(\alpha,E,E) - \frac{e^2}{2h}\hbar\omega' [\mathbf{s}^\dagger_{\alpha\beta}\partial_E \mathbf{s}_{\alpha\beta} - (\partial_E \mathbf{s}_{\alpha\beta})\mathbf{s}_{\alpha\beta}] \right\}, \tag{14}$$

where we have used the notation $A_{\alpha\beta} \equiv A_{\beta\beta}(\alpha,E,E)$ and $\mathbf{s}_{\alpha\beta} \equiv \mathbf{s}_{\alpha\beta}(E,U)$. The transmission function A is defined in the usual form as

$$A_{\beta\beta'}(\alpha,E,E',U) = \mathbf{1}_\alpha \delta_{\alpha\beta}\delta_{\alpha\beta'} - \mathbf{s}^\dagger_{\alpha\beta}(E,U)\mathbf{s}_{\alpha\beta'}(E',U). \tag{15}$$

In deriving Eq. (14), we have used the fact that $\Sigma_\beta A_{\beta\beta}(\alpha,E,E,U) = 0$. All quantities such as $A_{\alpha\beta}$, $\mathbf{s}_{\alpha\beta}$, and $u_\beta = [\partial U(\mathbf{r})/\partial V_\beta]_{eq}$ are taken at equilibrium, i.e., at $V_\alpha = 0$. Using Eq. (3), we separate the operator according to $\hat{O}_\beta^{(1)}\partial_E = V_\beta \partial_E + \Sigma_\gamma V_\gamma \partial_{V_\gamma}$, thus Eq. (14) can further be simplified to

$$I_\alpha(\omega') = \int dE(-\partial_E f)\Bigg(\sum_\beta V_\beta \frac{e}{h} A_{\alpha\beta} - \frac{e^2}{2h}\hbar\omega' \sum_\beta V_\beta [\mathbf{s}^\dagger_{\alpha\beta}\partial_E \mathbf{s}_{\alpha\beta} - (\partial_E \mathbf{s}_{\alpha\beta})\mathbf{s}_{\alpha\beta}] - \frac{e^2}{2h}\hbar\omega' \sum_\gamma V_\gamma \sum_\beta [\mathbf{s}^\dagger_{\alpha\beta}\partial_{V_\gamma}\mathbf{s}_{\alpha\beta} - (\partial_{V_\gamma}\mathbf{s}_{\alpha\beta})\mathbf{s}_{\alpha\beta}] \Bigg). \tag{16}$$

From this result, we immediately realize that the first term on the right-hand side is just the dc contribution to the electric current. From the second and third terms that are linear in ω' and V_β, we obtain the linear order emittance

$$E_{\alpha\beta} = \int dE(-\partial_E f)\frac{e^2}{4\pi i}\Bigg\{ [\mathbf{s}^\dagger_{\alpha\beta}\partial_E \mathbf{s}_{\alpha\beta} - (\partial_E \mathbf{s}_{\alpha\beta})\mathbf{s}_{\alpha\beta}] + \sum_\gamma [\mathbf{s}^\dagger_{\alpha\gamma}\partial_{V_\beta}\mathbf{s}_{\alpha\gamma} - (\partial_{V_\beta}\mathbf{s}_{\alpha\gamma})\mathbf{s}_{\alpha\gamma}] \Bigg\}. \tag{17}$$

This result exactly agrees with that obtained previously.[2] The first term of $E_{\alpha\beta}$ describes the external contribution to the ac current, while the second term is from internal response. They are obtained simultaneously from the scattering matrix theory developed here. Finally, from the gauge invariance condition (Ref. 2) $e\partial_E A_{\alpha\beta} + \Sigma_\gamma \partial_{V_\gamma} A_{\alpha\beta} = 0$, it is easy to show that $\Sigma_\beta E_{\alpha\beta} = 0$, which is a direct consequence of $\hat{O}_\beta^{(1)}$ in Eq. (14). It is also easy to show that the electric current is conserved, i.e., $\Sigma_\alpha E_{\alpha\beta} = 0$.

B. Third-order dc nonlinear conductance

The scattering matrix theory developed here can be applied to compute dc weakly nonlinear conductance to any order in bias. As an example we now calculate the third-order dc nonlinear conductance $G_{\alpha\beta\gamma\delta}$, which is defined by expanding the electric current in powers of voltage to the third power,

$$I_\alpha = \sum_\beta G_{\alpha\beta}V_\beta + \sum_{\beta\gamma} G_{\alpha\beta\gamma}V_\beta V_\gamma + \sum_{\beta\gamma\delta} G_{\alpha\beta\gamma\delta}V_\beta V_\gamma V_\delta + \cdots. \tag{18}$$

Following the same procedure as above in deriving the linear emittance $E_{\alpha\beta}$, by expanding the electric current Eq. (7) and other quantities to third order in bias, it is tedious but straightforward to derive

$$G_{\alpha\beta\gamma\delta} = \frac{e^3}{3h}\int dE(-\partial_E f)[\{\partial_{V_\gamma}\partial_{V_\delta}A_{\alpha\beta} + e\partial_{V_\gamma}\partial_E A_{\alpha\beta}\delta_{\beta\delta}\}_c + e^2\partial_E^2 A_{\alpha\beta}\delta_{\beta\gamma}\delta_{\gamma\delta}]. \tag{19}$$

Note that the second-order characteristic potential tensor has been implicitly included in Eq.(19), because

$$\partial_{V_\gamma}\partial_{V_\delta}A_{\alpha\beta}=\partial_{V_\gamma}\int \frac{\delta A_{\alpha\beta}}{\delta U(\mathbf{r}_1)}\frac{\partial U(\mathbf{r}_1)}{\partial V_\delta}d\mathbf{r}_1$$

$$=\int\left(\frac{\delta}{\delta U(\mathbf{r}_2)}\frac{\delta A_{\alpha\beta}}{\delta U(\mathbf{r}_1)}\right)u_\delta u_\gamma d\mathbf{r}_1 d\mathbf{r}_2$$

$$+\int \frac{\delta A_{\alpha\beta}}{\delta U(\mathbf{r}_1)}u_{\delta\gamma}d\mathbf{r}_1. \quad (20)$$

Again, we emphasize that other higher-order nonlinear conductances can be calculated in a similar fashion. In general, the nth order characteristic potential tensor is needed for the $(n+1)$th order nonlinear conductance. Finally, we point out that this result, Eq. (19), can in fact be obtained by expanding the following electric current expression to the third order in voltage, $I_\alpha=2e/h\Sigma_\beta\int dEf(E-E_F-eV_\beta)A_{\alpha\beta}(E,\{V_\gamma\})$.

In the following we calculate G_{1111} from the general result of Eq. (19) for a double-barrier tunneling diode. For simplicity let us consider a quasi-one-dimensional (1D) double-barrier tunneling system where the two barriers are δ functions located at positions $x=-a$ and $x=a$. The barrier strength is V_1 and V_2, respectively. They should be viewed as infinitely large planar barriers parallel to the y-z plane and transport is along the x direction. When $V_1=V_2$, this is a symmetric system, hence the second-order nonlinear conductance vanishes. In the symmetric case the first nonlinear coefficient comes from the third order, as specified by Eq. (19). If we approximate the scattering matrix by the Breit-Wigner form[8] near a resonance energy E_r, $s_{\alpha\beta}(E)\sim[\delta_{\alpha\beta}-i\sqrt{\Gamma_\alpha\Gamma_\beta}/\Delta]$, where Γ_α is the decay width of barrier α, $\Delta=\Delta E+i\Gamma/2$ with $\Gamma=\Gamma_1+\Gamma_2$ and $\Delta E\equiv E-E_r$, we obtain a simple expression,

$$G_{1111}=\frac{2e^3}{3h\Gamma^2}\left[\left(3(\Delta E)^2-\frac{\Gamma^2}{4}\right)(\Gamma_1^2+\Gamma_2^2-\Gamma_1\Gamma_2)\Gamma_1\Gamma_2 - 6\Gamma_1^2\Gamma_2^2(\Delta E)^2\right]\left[(\Delta E)^2+\frac{\Gamma^2}{4}\right]^{-3}. \quad (21)$$

For the symmetric case, this expression reduces to

$$G_{1111}=-\frac{e^3\Gamma_1^2}{6h}\frac{3(\Delta E)^2+\Gamma_1^2}{[(\Delta E)^2+\Gamma_1^2]^3}, \quad (22)$$

which is negative definite and has one minimum at $\Delta E=0$. Because of the simple nature of the scattering matrix within the Breit-Wigner form, a general electric current expression has been obtained:[8] $I_\alpha=I_\alpha(V_\beta)$. We have thus calculated G_{1111} from this exact I-V relation, and it agrees exactly with the result (22) that comes from Eq. (19).

Most practical transport problems cannot be solved analytically. It is thus very important to be able to solve them numerically. Indeed, a distinct merit of the scattering matrix theory presented here is that it allows numerical computation, e.g., Eqs. (17) and (19) can be numerically evaluated for explicit scattering potentials of a conductor. We only mention that the functional derivatives of the transmission function $A_{\alpha\beta}$ with respect to the potential landscape $U(\mathbf{r})$ as appeared in Eq. (19), the potential derivatives, and the partial local density of states, which is needed in the emittance calculation, can all be determined via the scattering Green's function.

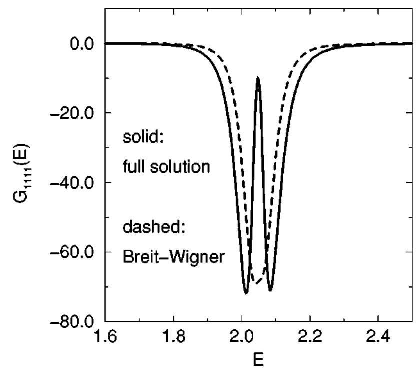

FIG. 1. G_{1111} as a function of the scattering electron energy E for a double barrier tunneling diode with symmetrical barriers. Solid line: numerical results by solving the full quantum scattering problem using Green's functions. Dashed curve: using the approximate Breit-Wigner form of the scattering matrix. The units of the quantities are set by $\hbar=1$, $e=1$, and $m=\frac{1}{2}$.

For the double-barrier diode just discussed, if we do not use the Breit-Wigner scattering form, G_{1111} can only be obtained numerically. For this system the Green's function $G(x,x')$ can be calculated exactly[16] thus from the Fisher-Lee relation[17] we obtain the scattering matrix $\mathbf{s}_{\alpha\beta}(E)$ and hence the transmission function $A_{\alpha\beta}$ from its definition (15). To compute $\delta A_{11}/\delta U$ and the higher-order functional derivative, we use Fisher-Lee relation and the fact[18] that $\delta G(x_1,x_2)/\delta U(x)=G(x_1,x)G(x,x_2)$. Hence,

$$\frac{\delta \mathbf{s}_{11}}{\delta U(x)}=i\hbar v G(x_1,x)G(x,x_1) \quad (23)$$

and

$$\frac{\delta^2\mathbf{s}_{11}}{\delta U(x)\,\delta U(x')}=i\hbar v[G(x_1,x')G(x',x)G(x,x_1) + G(x_1,x)G(x,x')G(x',x_1)], \quad (24)$$

where v is the velocity of the particle. The energy and V_γ derivatives of Eq. (19) can be evaluated explicitly using the numerical procedures documented before.[7,19] Finally, the nonlinear characteristic potential u_{11} is obtained from Eq. (12).

The numerical result for G_{1111} as a function of the scattering electron energy E is plotted as the solid curve in Fig. 1. Around the resonance energy $E_r\approx 2.0$, G_{1111} takes large but negative values. Thus for electron Fermi energy in this range, the current-voltage characteristics will be nonlinear and this may result to negative differential resistance. Such a behavior has important significance for practical purposes.[20] Notice that the two dips of G_{1111} are not exactly the same, such an asymmetrical behavior has been observed in other quantities.[16] For comparison we also plotted (dashed line) the result from the Breit-Wigner approximation of the scattering matrix, Eq. (22). While the negative nature of G_{1111} and the overall magnitude are similar to the numerical result, the Breit-Wigner result shows only one dip. This inconsistency is completely due to the simple form of the Breit-Wigner approximation as it gives a space independent and constant characteristic potential. The accurate solution using the Green's function generates space-dependence of various quantities.

IV. SUMMARY

In summary, we have extended the scattering matrix theory that is now appropriate to analyzing linear dynamic conductance to first order in frequency, and weakly nonlinear dc conductance order by order in external bias. The crucial ingredient of this development is the characteristic potential at weakly nonlinear orders and these potentials appear naturally from the self-consistent Hamiltonian. The theory is current conserving and gauge invariant. The physical quantities involved in this theory are numerically calculable, hence the present approach can be used to conductors with complicated scattering potential landscape for quantitative predictions. The formal connection of the scattering matrix theory and the response theory, at the weakly nonlinear ac level, has been clarified in our recent work.[14] While the response theory is very general[14] and can be used to analyze weakly nonlinear dc and ac transport order by order in bias, and for ac case to all orders of frequency, we expect that the scattering matrix theory should be able to do the same. This paper partially fulfills this expectation by extending the scattering matrix theory to higher orders of nonlinearity. We point out that to go beyond the linear frequency, the expression for electric current, Eq. (7), should be extended. In addition we should use the Helmholtz equation (in Lorentz gauge) for the electrodynamics instead of the Poisson equation. It is also important to note that the theoretical approach of scattering matrix developed here is appropriate to transport problems near equilibrium. Far from equilibrium, one may employ the Keldysh Green's functions.[21] In addition, the electron-electron interaction, which is needed in order to maintain the gauge invariance at the nonlinear level, is treated in the density-functional sense. Hence the present theory is applicable for situations where interactions can be analyzed using random-phase approximation type approach. This is, in fact, the typical situation for 2D and 3D mesoscopic conductors.

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