Nonlinear dynamic conductance and harmonic generation in mesoscopic multiprobe systems

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We report a theoretical analysis of nonlinear dynamic conductance and harmonic generation for multiprobe mesoscopic conductors. Our calculation takes into account the internal induced potential which is due to a response to oscillating external bias voltages applied at the probes of the conductor. We pay special attention to the physical requirement of current conservation and gauge invariance. There are generally several branches for higher-frequency harmonics in nonlinear components of the transport current, and each branch has its characteristic harmonics frequency. Our theory permits weakly nonlinear analysis order by order in terms of the external bias but in general terms of the ac frequency, and in particular we present detailed derivations up to third order. The third-order response contains a contribution at the driving frequency and a contribution 3 times the driving frequency. As a specific example we analyzed the nonlinear harmonic generation in double-barrier tunneling systems.

I. INTRODUCTION

The technological advance in fabricating nanostructures and the recent interest in physics of ultrasmall semiconductor devices have motivated formulations of appropriate quantum transport theories applicable for coherent multiprobe quantum conductors.[1,2] In particular, nonlinear quantum transport properties of these mesoscopic conductors have received much attention both theoretically and experimentally.[3,4] On the theoretical side, several approaches have been proposed to analyze quantum transport problems under dc and ac conditions, including the scattering matrix theory,[2,5] response theory,[6–9] nonequilibrium Green's function theory,[10–13] and direct numerical simulations. In the ballistic regime under an external dc bias, electronic transport can be described in terms of independent conducting channels characterized by their transmission coefficients[2,14] through the mesoscopic conductor. In a typical analysis of the linear dc conductance coefficient, which is the linear order coefficient of electric current versus bias voltage, it is *qualitatively not* essential to include the potential buildup inside the conductor: many examples have shown that single-electron scattering matrix theory is adequate in predicting the dc linear conductance of mesoscopic conductors.[1]

On the other hand, when comes to predicting nonlinear transport coefficients and ac transport coefficients for mesoscopic conductors, it is both qualitatively and quantitatively important to include a self-consistent internal potential caused by the long-range Coulomb interactions.[2] The reason is that the induced internal potential can be quite substantial for small conductors due to their small density of states which in turn leads to the long-screening lengths. Hence the total potential of the system, external plus induced, can be very different from the applied external potential. Furthermore, it has been shown[2] that without consideration of the induced potential, the theory would violate electric current conservation under ac bias and would violate gauge invariance under nonlinear dc situations. A major progress in developing viable nonlinear dc and ac theories has been achieved recently after Levinson[15] and Büttiker[16] introduced a self-consistent internal potential in the context of considering current conservation and gauge invariance requirements. Physically for ac transport situations, the self-consistent potential is induced by a redistribution of charges due to the presence of a displacement current when a time-dependent bias is applied to the conductor. Essentially, in response to a potential variation at a contact of the mesoscopic conductor, the charge distribution in the interior of the sample is driven away from equilibrium.[9,17] On the other hand, an electron Coulomb interaction opposes this variation. The competition of these two effects gives rise to a self-consistent internal potential which must be taken into account in predicting

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transport coefficients. Usually we can write the net charge, which is associated with the redistribution of carriers in the sample in response to an external bias potential variation, into two parts. The first part is due to direct external injection of carriers through contacts of the conductor with a fixed internal potential. The second part is due to induction which accounts for such effects as the displacement current, and which is sensitive to the density of states of the conductor. The effects of displacement current have been indicated by physical considerations[18] and by direct numerical simulations.[19]

When dealing with nonlinear ac response in quantum transport, higher-order harmonic generation is naturally expected, similar to those observed in nonlinear optical systems. In other words, due to nonlinearity, an external ac perturbation at frequency Ω can generate an electric current response at frequencies Ω and multiples of Ω including a dc component.[21] The dc component of harmonic generation has been analyzed recently. de Vegvar[20] studied the low-frequency second-harmonic transport response of mesoscopic conductors using a perturbation theory and he found that the low-frequency second-harmonic current is a non-Fermi-surface quantity. In a recent study Pedersen and Büttiker[21] discussed the effects of displacement current on harmonic generation. The current in the nonlinear regime is also studied on a double-barrier tunneling diode by Blanter and Büttiker.[22] These investigations were based on perturbation expansions in terms of both external bias and in frequency; hence the full frequency-dependent feature has not been clearly examined. It is the purpose of this work to report a systematic theoretical analysis on nonlinear ac transport where harmonic generation is studied in general terms of the ac frequency.

To make the problem of harmonic generation clearer, let us consider the weakly nonlinear response of a mesoscopic conductor to an applied external bias voltage. For this situation the electric current flowing through a probe labeled by α, $I_\alpha(t)$, can be expanded in powers of the bias voltage $V_\beta(t)$ applied at probe β. V_β is finite but is assumed to be small; hence the weakly nonlinear expansion makes sense and only the first few terms need to be retained. The expansion is written as

$$\begin{aligned} I_\alpha(t) = &\sum_\beta \int_{t_0}^{t} dt_1 G_{\alpha\beta\gamma}(t-t_1) V_\beta(t_1) \\ &+ \sum_{\beta\gamma} \int_{t_0}^{t} dt_1 \int_{t_0}^{t} dt_2 G_{\alpha\beta\gamma}(t_1-t_2, t-t_2) V_\beta(t_1) V_\gamma(t_2) \\ &+ \sum_{\beta\gamma\delta} \int_{t_0}^{t} dt_1 \int_{t_0}^{t} dt_2 \int_{t_0}^{t} dt_3 \\ &\times G_{\alpha\beta\gamma\delta}(t_2-t_3, t_1-t_3, t-t_3) V_\beta(t_1) V_\gamma(t_2) V_\delta(t_3) \\ &+ \cdots . \end{aligned} \tag{1}$$

Here it has been assumed that the bias voltage at probe α is turned on at time t_0. The response coefficients $G_{\alpha\beta\gamma}(t-t_1)$, $G_{\alpha\beta\gamma}(t_1-t_2,t-t_2)$, and $G_{\alpha\beta\gamma\delta}(t_2-t_3,t_1-t_3,t-t_3)$ are, respectively, the first-, second-, and third-order dynamic conductance. To make the analysis relatively easier, in the following discussions we assume $V_\alpha(t)$ to be a simple oscillating form turned on adiabatically at $t_0=-\infty$, i.e., $V_\alpha(t) = V_\alpha e^{-\eta|t|}\cos\Omega t$, where η is positive and infinitesimal. The general form of the various terms in the expansion (1) can be found by invoking the fundamental physical principle of time invariance: the dynamical properties of our system cannot be changed by a translation of the time origin. For our case, a time displacement of external bias merely results in a corresponding time displacement of the induced current. By changing variables in the time integration of Eq. (1), one has

$$\begin{aligned} I_\alpha(t) = &\mathrm{Re}\left[e^{i(\Omega-i\eta)t} \sum_\beta G^{(\Omega)}_{\alpha\beta}(\Omega) V_\beta \right] \\ &+ \frac{1}{2} e^{2\eta t} \sum_{\beta\gamma} G^{(0)}_{\alpha\beta\gamma}(\Omega) V_\beta V_\gamma \\ &+ \frac{1}{2} \mathrm{Re}\left[e^{i2(\Omega-i\eta)t} \sum_{\beta\gamma} G^{(2\Omega)}_{\alpha\beta\gamma}(\Omega) V_\beta V_\gamma \right] \\ &+ \frac{1}{2^2} \mathrm{Re}\left[e^{i(\Omega-i\eta)t} e^{2\eta t} \sum_{\beta\gamma\delta} G^{(\Omega)}_{\alpha\beta\gamma\delta}(\Omega) V_\beta V_\gamma V_\delta \right] \\ &+ \frac{1}{2^2} \mathrm{Re}\left[e^{i3(\Omega-i\eta)t} \sum_{\beta\gamma\delta} G^{(3\Omega)}_{\alpha\beta\gamma\delta}(\Omega) V_\beta V_\gamma V_\delta \right], \end{aligned} \tag{2}$$

where Re[ ] means the real part of [ ]. In this expression of the current, $G^{(\Omega)}_{\alpha\beta}(\Omega) \equiv \int_0^\infty d\tau_1 G_{\alpha\beta}(\tau_1) e^{-i(\Omega-i\eta)\tau_1}$ is the linear complex dynamic conductance coefficient, called admittance,

$$\begin{aligned} G^{(0)}_{\alpha\beta\gamma}(\Omega) = &\int_0^\infty d\tau_1 \int_0^\infty d\tau_2 G_{\alpha\beta\gamma} \\ &\times (\tau_2, \tau_1+\tau_2) e^{-\eta(2\tau_1+\tau_2)} \cos\Omega\tau_2 \end{aligned}$$

and

$$G^{(2\Omega)}_{\alpha\beta\gamma}(\Omega) = \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 G_{\alpha\beta\gamma}(\tau_2, \tau_1+\tau_2) e^{-i(\Omega-i\eta)(2\tau_1+\tau_2)}$$

are second-order complex dynamic conductance coefficients, and

$$\begin{aligned} G^{(\Omega)}_{\alpha\beta\gamma\delta}(\Omega) = &\int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3 G_{\alpha\beta\gamma\delta} \\ &\times (\tau_3, \tau_2+\tau_3, \tau_1+\tau_2+\tau_3) \\ &\times [2e^{-\eta(2\tau_1+2\tau_2+\tau_3)} e^{-i(\Omega-i\eta)\tau_1} \cos\Omega\tau_3 \\ &+ e^{-2\eta\tau_1} e^{-i(\Omega-i\eta)(\tau_1+2\tau_2+\tau_3)}] \end{aligned}$$

and

$$\begin{aligned} G^{(3\Omega)}_{\alpha\beta\gamma\delta}(\Omega) = &\int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3 G_{\alpha\beta\gamma\delta} \\ &\times (\tau_3, \tau_2+\tau_3, \tau_1+\tau_2+\tau_3) \\ &\times e^{-i3(\Omega-i\eta)(\tau_1+\tau_2+\tau_3)} \end{aligned}$$

are third-order complex dynamic conductance coefficients.

Equation (2) indicates clearly that the second-order term in the electric current contains a dc piece and an oscillating piece at twice the driving frequency Ω. In addition Eq. (2) indicates that at third order there is a piece oscillating at the driving frequency and a piece oscillating at triple the driving frequency. These together with even higher-order terms give the harmonic generation efficiency. The nonlinear terms of Eq. (2) suggest that in addition to the component oscillating at the driving frequency Ω, the current contains significant components oscillating at harmonic frequencies 2Ω, 3Ω, . . . plus a dc component at zero frequency. This is precisely analogous to the well-known harmonic distortion of signals in an electrical circuit whose dynamic response is not perfectly linear.

In this paper we will investigate harmonic generation and its corresponding ac dynamic conductance coefficients up to third order in an external bias. Our calculations concern not only dc transport features but also frequency-dependent features in weakly nonlinear response. The derived analytical expressions presented in this paper are suitable for predicting ac transport coefficients in general terms of the frequency, rather than in a form expanded in frequency as done in previous studies. Our investigation is carried out by combining response theory[7] with scattering matrix theory:[2] in particular we generalize the scattering matrix approach to take into account nonlinear dependences on oscillating potentials. For the most general case of a mesoscopic conductor under external ac fields, a theoretical analysis is, perhaps, impossible to be carried out if full electrodynamic effects such as Faraday's law need to be included. We thus neglect this secondary effect and only include the dynamic electric field response. We however point out that, formally, the full Maxwell equations can indeed be included into our formalism, but that makes analytical derivations very difficult[9] and is beyond the scope of this work. Our analysis indicates that there is only one branch in the linear response while there are more than one branches of transport current in the nonlinear response: each branch has its own frequency characteristics as discussed above, Eq. (2). Importantly, the ac external perturbation produces static dc components in the current or a dc electric field in the conductor; i.e., there is an optical rectification effect at nonlinear orders.

As mentioned above, at nonlinear order special attention must be paid to gauge invariance: the electric current cannot depend on the choice of potential zero. In other words, $I_\alpha(t)$ of Eq. (1) cannot change if all the bias voltages are shifted by the same constant: the value of I_α remains the same when $V_\beta \rightarrow V_\beta + \Delta$ for all β. In addition, current conservation means $\Sigma_\alpha I_\alpha(t)=0$. These requirements, applied to Eq. (1), suggest that dynamic conductance coefficients must satisfy many sum rules, $\Sigma_\alpha G^{(n\Omega)}_{\alpha\beta\cdots\delta}(\omega)=\Sigma_\beta G^{(n\Omega)}_{\alpha\beta\cdots\delta}(\omega)=\cdots=\Sigma_\delta G^{(n\Omega)}_{\alpha\beta\cdots\delta}(\omega)=0$, where $n>1$ is an integer indicating the order of harmonic generation. Our theory, to be presented below, produces dynamic conductance coefficients satisfying these constraints.

The rest of paper is organized as follows: in Sec. II the current response to an external ac bias is then calculated perturbatively. By separating harmonics, the corresponding dynamic conductance coefficients are derived: we explicitly confirm current conservation and gauge invariance under the Thomas-Fermi approximation. In Sec. III we apply our formalism to analyze double-barrier tunneling systems. The last section contains a short summary and related discussions. There are two appendixes: In Appendix A we gave the derivation of the Hamiltonian with considerations of external applied voltage and internal induced potential. The solution is obtained by solving the equation of motion. In Appendix B the essential derivations of charge density as well as the equations for the characteristic potential tensors have been presented.

II. THEORY

Our analysis is based on a perturbation theory, scattering matrix theory, and a self-consistent solution of the internal potential built up at the Hartree level. To make the presentation clear we put detailed mathematical derivations inside two appendixes. First, by iterating the equation of motion we derive the correlation function which enters the electric current operator; second, we derive equations satisfied by the internal potential buildup in terms of the local density of states (LDOS); combining these results we obtain the dynamic conductance coefficients.

The system considered here is a small volume of the multiprobe sample with an application of the time-dependent voltages $V_\alpha(t)$ varying with frequency Ω at the probes α. We suppose that the volume is sufficiently small so that the spatial variation of electric field could be ignored. Also the effects arising from the induced magnetic field are ignored. Following the works of Büttiker and co-workers,[2,16] the current flowing over the probe α is found:[2]

$$I_\alpha(t)=\frac{e}{h}\int dE\sum_{\beta k} A_{\beta\beta,kk}(\alpha,E,E')\langle C^\dagger_{\beta k}(E,t)C_{\beta k}(E',t)\rangle_{eq}, \tag{3}$$

where

$$A_{\beta\beta,kk}(\alpha,E,E')=\sum_l \left[\mathbf{1}_\alpha\delta_{\alpha\beta}\delta_{kl}-S^\dagger_{\alpha\beta,kl}(E,U(\mathbf{r},\{V\}))\times S_{\alpha\beta,kl}(E',U(\mathbf{r},\{V\}))\right] \tag{4}$$

is the screened transmission function in terms of scattering matrix $S_{\alpha\beta,kl}(E,U(\mathbf{r},\{V\}))$, which is a functional of energy E and induced internal potential $U(\mathbf{r},\{V_\alpha\})$. $C_{\beta k}$ ($C^\dagger_{\beta k}$) is the annihilation (creation) operator for an electron in the incoming channel k inside the probe β and the scattering matrix is used to describe the relationship between the incoming electron at the probe β to the outgoing electron at the probe α. From Eq. (3), therefore, the current can be obtained upon the solution $C^\dagger_{\beta k}(E,t)C_{\beta k}(E't)$ being known. This double operator can be found by solving the equation of motion perturbatively.

The Hamiltonian can be written in the following form:

$$H=\sum_{\alpha\beta,mn}\int dEdE'E'\left[\delta_{\alpha\beta}\delta_{mn}\delta(E-E')+\frac{e}{2\pi i}\mathcal{B}_{\alpha\beta,mn}(E,E',t)\right]C^\dagger_{\alpha m}(E)C_{\beta n}(E'), \tag{5}$$

where $\mathcal{B}_{\alpha\beta,mn}(E,E',t)=\tilde{V}_\alpha(t)\delta_{\alpha\beta}\delta_{mn}(E-E')^{-1}$ and $e\tilde{V}_\alpha(t)=eV_\alpha+\int d\mathbf{r}[\delta E_\alpha(U)/\delta U]U(\mathbf{r},t)$ is the global voltage at the probe α in which the effects of internal potential have been included. The second term in the Hamiltonian is regarded as a time-dependent perturbation $H_I(t)$ because of the function $e^{-\eta|t|}\cos\Omega t$ in the applied voltages, which satisfies $H_I(-\infty)=0$. The derivation of Eq. (5) is given in Appendix A. As shown in the global voltages $\tilde{V}_\alpha$ the total perturbation to which the electron system responds is the sum of applied perturbation and internal potentials due to the induced variation of the electron density. The internal potential is determined by Poisson's equation. In Appendix B, we express the internal potential in terms of characteristic potentials and obtain the equations for these characteristic potentials.

Using the time-evolution equation for a double operator $\mathbf{Q}_{\alpha\beta,kk}(E,E',t)=C^\dagger_{\alpha k}(E,t)C_{\beta k}(E',t)$,

$$i\hbar\partial_t\mathbf{Q}_{\alpha\beta,kk}(E,E',t)=[\mathbf{Q}_{\alpha\beta,kk}(E,E',t),H], \tag{6}$$

and employing the equal-time commutation relations

$$\{C_{\alpha m}(E,t),C^\dagger_{\beta n}(E',t)\}=\delta_{\alpha\beta}\delta_{mn}\delta(E-E')$$

and

$$\{C_{\alpha m}(E,t),C_{\beta n}(E',t)\}=\{C^\dagger_{\alpha m}(E,t),C^\dagger_{\beta n}(E',t)\}=0,$$

one obtains

$$\begin{aligned}i\hbar\partial_t\mathbf{Q}_{\alpha\alpha,kk}(E,E',t)&=(E'-E)\mathbf{Q}_{\alpha\alpha,kk}(E,E',t)\\&\quad+\frac{e}{2\pi i}\sum_\beta\int dE''[\mathcal{B}_{\beta\alpha,nk}(E',E'',t)\\&\quad\times\mathbf{Q}_{\alpha\beta,kk}(E,E'',t)\\&\quad-\mathcal{B}_{\alpha\beta,kn}(E'',E,t)\mathbf{Q}_{\alpha\beta,kk}(E'',E',t)].\end{aligned} \tag{7}$$

Making use of a transformation $\tilde{C}^\dagger_{\alpha k}(E,t)\tilde{C}_{\alpha k}(E',t) = \exp[(i/\hbar)(E'-E)t]C^\dagger_{\alpha k}(E,t)C_{\alpha k}(E',t)$ and integrating the equation of motion with respect to the time variable, then going back to $C^\dagger_{\alpha k}(E,t)C_{\alpha k}(E',t)$, it is found that

$$\begin{aligned}C^\dagger_{\alpha k}(E,t)C_{\alpha k}(E',t)=e^{-i(E'-E)t/\hbar}\Bigg\{&C^\dagger_{\alpha k}(E)C_{\alpha k}(E')+\frac{1}{i\hbar}\frac{e}{2\pi i}\int_{-\infty}^t dt_1\sum_{\beta,n}e^{i(E'-E)t_1/\hbar}[\mathcal{B}_{\beta\alpha,nk}(E',E'',t_1)\\&\times C^\dagger_{\alpha k}(E,t_1)C_{\beta n}(E'',t_1)-\mathcal{B}_{\alpha\beta,kn}(E'',E,t_1)C^\dagger_{\beta n}(E'',t_1)C_{\alpha k}(E',t_1)]\Bigg\}.\end{aligned} \tag{8}$$

The perturbation solution up to third order in powers of $\mathcal{B}_{\alpha\beta,mn}(E,E',t)$ is presented in Appendix A.

Substituting the solution of couple operator (A9) into Eq. (3), the current can be calculated in a standard way. Here we would like to comment that in evaluating the current, it is needed to take a quantum statistical average of $\langle C^\dagger_{\alpha k}(E)C_{\beta k}(E')\rangle_{eq}=\delta_{\alpha\beta}\delta(E-E')f_\alpha(E)$, where $f_\alpha(E)$ is the Fermi function of reservoir α. It is assumed that the modulation imposed on the system is so slow that the contacts can still be regarded as being in a dynamic equilibrium state. Thus the quantum statistical average can be found by evaluating the averages of $C_{\alpha k}(E)$. From Eq. (A9), one obtains

$$\begin{aligned}\langle C^\dagger_{\alpha k}(E,t)C_{\alpha k}(E',t)\rangle_{eq}&\\=f_\alpha(E)\delta(E-E')+\mathcal{W}_1(t)&\\+\mathcal{W}_2(t)+\mathcal{W}_3(t),&\end{aligned} \tag{9}$$

where $\mathcal{W}_j(E,t)$ are given in Eqs. (A13)–(A15) in powers of the global voltages $\tilde{V}_\beta(t)$. According to Eq. (B3) and considering a series expansion of energy in terms of potential landscape U, the global voltages can be written in powers of the applied voltages

$$\tilde{V}_\beta(t)=\sum_{j=1}V^{(j)}_\beta h_j(t), \tag{10}$$

where the linear, the second-order, and the third-order modified voltages at the probe β are

$$V^{(1)}_\beta=\sum_\gamma V_\gamma\left(\delta_{\gamma\beta}+\int d\mathbf{r}u_\gamma(\mathbf{r})\frac{\delta E(U)}{\delta U(\mathbf{r})}\right), \tag{11}$$

$$V^{(2)}_\beta=\frac{1}{2}e\sum_{\gamma\delta}V_\gamma V_\delta\int d\mathbf{r}u_{\gamma\delta}(\mathbf{r})\frac{\delta E(U)}{\delta U(\mathbf{r})}, \tag{12}$$

$$V^{(3)}_\beta=\frac{1}{3!}e^2\sum_{\gamma\delta\rho}V_\gamma V_\delta V_\rho\int d\mathbf{r}u_{\gamma\delta\rho}(\mathbf{r})\frac{\delta E(U)}{\delta U(\mathbf{r})}, \tag{13}$$

and $h_j(t)=\cos^j\Omega te^{-\eta|t|}$ (j=1, 2, and 3) are the time-dependent factors.

The first term $f(E)\delta(E-E')$ in Eq. (9) does not contribute to the transport electric current. In equilibrium, the current containing this term is $I^{(0)}_\alpha=\Sigma_{\beta k}f(E)A_{\beta\beta,kk}(\alpha,E,E)$, which vanishes immediately due to $\Sigma_{\beta k}A_{\beta\beta,kk}(\alpha,E,E)=0$. It corresponds to the closed current loops following the equipotential contours when the magnetic field is applied to the system. We will neglect this term in our further calculations.

To view the frequency dependence of current, we transform $\langle C^\dagger_{\alpha k}(E,t)\ C_{\alpha k}(E',t)\rangle_{eq}$, in Eq. (9), into its Fourier transformation form. In addition, the Fourier transformation forms of globe voltages become $\tilde{V}_\beta(\nu)=\Sigma_{j\gamma}V^{(j)}_\gamma(\beta)h_j(\nu)$, where the frequency-dependent factor $h_j(\nu)$ is the Fourier transformation of $h_j(t)$,

$$h_1(\nu)=\frac{1}{2}[\delta(\nu+\Omega)+\delta(\nu-\Omega)], \tag{14}$$

$$h_2(\nu)=\frac{1}{2^2}[\delta(\nu+2\Omega)+\delta(\nu-2\Omega)+2\delta(\nu)], \tag{15}$$

and

$$h_3(\nu)=\frac{1}{2^3}[\delta(\nu+3\Omega)+\delta(\nu-3\Omega)+3\delta(\nu+\Omega)+3\delta(\nu-\Omega)]. \tag{16}$$

Then the Fourier transformation form of current, Eq. (3), is obtained:

$$I_\alpha(\omega)=\frac{e}{h}\int dEdE'\sum_{\beta k} A_{\beta\beta,kk}(\alpha,E,E')\times[\mathcal{W}_1(\omega)+\mathcal{W}_2(\omega)+\mathcal{W}_3(\omega)], \tag{17}$$

where $\mathcal{W}_j(\omega)=(2\pi)^{-1}\int_{-\infty}^{\infty}dte^{i\omega t}\mathcal{W}_j(t)$ is the Fourier transformation of $\mathcal{W}_j(t)$. With the help of Eqs. (A13)–(A15), it is found that

$$\mathcal{W}_1(\omega)=-\frac{e}{2\pi i}\sum_{\gamma}\tilde{V}_\beta(\omega)\frac{f(E)-f(E')}{E-E'}\frac{1}{E-E'+\hbar\omega+i\eta},$$

$$\mathcal{W}_2(\omega)=-\frac{1}{2!}\frac{e^2}{(2\pi i)^2}\int_{-\infty}^{\infty}d\nu\tilde{V}_\beta(\nu)\tilde{V}_\beta(\omega-\nu)\int dE''\frac{1}{E-E'+\hbar\omega+i\eta}\times\Bigg[[(f(E'')-f(E)]\frac{1}{E'-E''}\frac{1}{E''-E}\times\frac{1}{E-E''+\hbar(\omega-\nu)+i\eta}+[f(E'')-f(E')]\frac{1}{E'-E''}\frac{1}{E''-E}\frac{1}{E''-E'+\hbar(\omega-\nu)+i\eta}\Bigg],$$

and

$$\begin{aligned}\mathcal{W}_3(\omega)=&-\frac{1}{3!}\frac{e^3}{(2\pi i)^3}\int_{-\infty}^{\infty}\nu_1\int_{-\infty}^{\infty}d\nu_2\tilde{V}_\beta(\nu_1)\tilde{V}_\beta(\nu_2)\tilde{V}_\beta(\omega-\nu_1-\nu_2)\int dE''dE'''\frac{1}{E-E'+\hbar\omega+i\eta}\\&\times\Bigg\{[f(E)-f(E'')]\frac{1}{E'''-E''}\frac{1}{E''-E}\frac{1}{E'-E'''}\frac{1}{E-E''+\hbar(\omega-\nu_1-\nu_2)+i\eta}\frac{1}{E-E'''+\hbar(\omega-\nu_1)+i\eta}\\&-[f(E'')-f(E''')]\frac{1}{E'''-E''}\frac{1}{E''-E}\frac{1}{E'-E'''}\frac{1}{E''-E'''-\hbar(\omega-\nu_1-\nu_2)+i\eta}\frac{1}{E-E'''+\hbar(\omega-\nu_1)+i\eta}\\&-[f(E''')-f(E'')]\frac{1}{E'-E''}\frac{1}{E''-E'''}\frac{1}{E'''-E}\frac{1}{E'''-E''+\hbar(\omega-\nu_1-\nu_2)+i\eta}\frac{1}{E'''-E'+\hbar(\omega-\nu_1+i\eta)}\\&+[f(E'')-f(E')]\frac{1}{E'-E''}\frac{1}{E''-E'''}\frac{1}{E'''-E}\frac{1}{E''-E'+\hbar(\omega-\nu_1-\nu_2)+i\eta}\frac{1}{E'''-E'+\hbar(\omega-\nu_1)+i\eta}\Bigg\}.\end{aligned}$$

In the calculation of these integrations of energy, there is a relation

$$\int_{-\infty}^{\infty}dE'X(E')\frac{1}{E-E'+i\eta}=2\pi iX(E) \tag{18}$$

for an arbitrary function $X(E)$. Therefore, integrating over E' in Eq. (17), the Fourier transformation form of the current is then written as

$$I_\alpha(\omega)=\frac{e}{h}\int dE\sum_{\beta k} A_{\beta\beta,kk}(\alpha,E,E+\hbar\omega)\langle C^\dagger_{\beta k}(E)C_{\beta k}(E+\hbar\omega)\rangle_{eq}, \tag{19}$$

where the quantum statistical average $\langle C^\dagger_{\beta k}(E)C_{\beta k}(E+\hbar\omega)\rangle_{eq}$ is given by the following relation:

$$\langle C^{\dagger}_{\beta k}(E)C_{\beta k}(E+\hbar\omega)\rangle_{eq}=\frac{e}{2}\left[\sum_{j=1}^{3}V_{\beta}^{(j)}\frac{h_j(\omega)}{\hbar\omega}\right][f(E)-f(E+\hbar\omega)]$$
$$-\frac{e^2}{2!}V_{\beta}^{(1)}V_{\beta}^{(1)}\int_{-\infty}^{\infty}d\nu\frac{h_1(\nu)}{\hbar\nu}\frac{h_1(\omega-\nu)}{\hbar(\omega-\nu)}[f(E+\hbar(\omega-\nu_1))-f(E)+f(E+\hbar\nu)-f(E+\hbar\omega)]$$
$$-\frac{e^2}{2!}V_{\beta}^{(1)}V_{\beta}^{(2)}\int_{-\infty}^{\infty}d\nu\left[\frac{h_1(\nu)}{\hbar\nu}\frac{h_2(\omega-\nu)}{\hbar(\omega-\nu)}+\frac{h_1(\omega-\nu)}{\hbar(\omega-\nu)}\frac{h_2(\nu)}{\hbar\nu}\right][f(E+\hbar(\omega-\nu_1))-f(E)+f(E+\hbar\nu)$$
$$-f(E+\hbar\omega)]+\frac{e^3}{3!}V_{\beta}^{(1)}V_{\beta}^{(1)}V_{\beta}^{(1)}\int_{-\infty}^{\infty}d\nu_1\int_{-\infty}^{\infty}d\nu_2\frac{h_1(\nu_1)}{\hbar\nu_1}\frac{h_1(\nu_2)}{\hbar\nu_2}\frac{h_1(\omega-\nu_1-\nu_2)}{\hbar(\omega-\nu_1-\nu_2)}$$
$$\times[f(E)-f(E+\hbar(\omega-\nu_1-\nu_2))-f(E+\hbar\nu_2)+f(E+\hbar(\omega-\nu_1))-f(E+\hbar\nu_1)$$
$$+f(E+\hbar(\omega-\nu_2))+f(E+\hbar(\nu_1+\nu_2))-f(E+\hbar\omega)]. \tag{20}$$

Furthermore, from Eqs. (14)–(16), it is easy to find

$$h_1(\nu)h_1(\omega-\nu)=\frac{1}{2^2}\left\{\delta\left(\nu-\frac{\omega}{2}\right)[\delta(\omega+2\Omega)+\delta(\omega-2\Omega)]+\delta(\omega)[\delta(\nu+\Omega)+\delta(\nu-\Omega)]\right\}, \tag{21}$$

$$h_1(\nu)h_2(\omega-\nu)=\frac{1}{2^3}\left\{\delta\left(\nu-\frac{\omega}{3}\right)[\delta(\omega+3\Omega)+\delta(\omega-3\Omega)]+[\delta(\nu+\omega)+2\delta(\nu-\omega)][\delta(\omega+\Omega)+\delta(\omega-\Omega)]\right\}, \tag{22}$$

and

$$h_2(\nu)h_1(\omega-\nu)=\frac{1}{2^3}\left\{\delta\left(\nu-\frac{2\omega}{3}\right)[\delta(\omega+3\Omega)+\delta(\omega-3\Omega)]+[\delta(\nu-2\omega)+2\delta(\nu)]\times[\delta(\omega+\Omega)+\delta(\omega-\Omega)]\right\}. \tag{23}$$

After expanding the frequency-dependent expression to first order in frequency ω with the consideration of a low-frequency nonlinear response, the transport electric current is finally expressed in the following form:

$$I_{\alpha}(\omega)=[\delta(\omega+\Omega)+\delta(\omega-\Omega)]I_{\alpha}^{(1)}(\omega)+\delta(\omega)I^{(2)}(0)+[\delta(\omega+2\Omega)+\delta(\omega-2\Omega)]I^{(2)}(\omega)+\{[\delta(\omega+3\Omega)+\delta(\omega-3\Omega)]+3[\delta(\omega+\Omega)+\delta(\omega-\Omega)]\}I^{(3)}(\omega), \tag{24}$$

where

$$I_{\alpha}^{(1)}(\omega)=-\frac{e^2}{2h}\int dE\sum_{\beta}V_{\beta}^{(1)}\mathcal{F}(E,\omega)\mathcal{A}_{\alpha\beta}(E,\omega), \tag{25}$$

$$I^{(2)}(\omega)=-\frac{e^2}{2^2h}\int dE\sum_{\beta}\left[V_{\beta}^{(2)}\mathcal{F}(E,\omega)-\frac{e}{2}V_{\beta}^{(1)}V_{\beta}^{(1)}\frac{\partial\mathcal{F}(E,\omega)}{\partial E}\right]\mathcal{A}_{\alpha\beta}(E,\omega), \tag{26}$$

and

$$I^{(3)}(\omega)=-\frac{e^2}{2^3h}\int dE\sum_{\beta}\left[V_{\beta}^{(3)}\mathcal{F}(E,\omega)-eV_{\beta}^{(1)}V_{\beta}^{(2)}\frac{\partial\mathcal{F}(E,\omega)}{\partial E}+\frac{e^2}{3!}V_{\beta}^{(1)}V_{\beta}^{(1)}V_{\beta}^{(1)}\frac{\partial^2\mathcal{F}(E,\omega)}{\partial E^2}\right]\mathcal{A}_{\alpha\beta}(E,\omega), \tag{27}$$

with $\mathcal{F}(E,\omega)=f'(E)+\frac{1}{2}\hbar\omega f''(E)$ and $\mathcal{A}_{\alpha\beta}(E,\omega)=\mathrm{Tr}\,A_{\beta\beta}(\alpha,E,E+\hbar\omega)$. ''Tr'' is the trace taken over the channel index k. In view of the frequency-dependent factors in this expression, the harmonic generation is found explicitly. In the linear response, there is only one branch of transport current with the driving frequency. However, in the nonlinear response, there are several branches for each order of the expansion. There are two branches for the second-order response functions. One is dc current; the corresponding static term produces a dc electric field in the conductor, which called an optical rectification effect. Beyond this dc branch, there is a response oscillating at twice the applied frequency, which gives rise to second-harmonic generation. The same thing happens in the higher-order response functions. The third-order response current contains a contribution representing the oscillation at the usual driving frequency and a contribution representing the oscillation at triple the driving frequency. The latter gives rise to third-harmonic generation.

The generalized conductances are defined as the derivatives of the transport electric current with respect to the voltages, $G_{\alpha\beta\cdots\{j\}}(\omega)=[d^jI_{\alpha}(\omega)/dV_{\beta}\cdots dV_{\{j\}}]|_{\{V\}=0}$. With the help of Eq. (24), one finds the linear conductance

$$G_{\alpha\beta}(\omega)=[\delta(\omega+\Omega)+\delta(\omega-\Omega)]G^{(\Omega)}_{\alpha\beta}(\omega), \tag{28}$$

with

$$G^{(\Omega)}_{\alpha\beta}(\omega)=\frac{e^2}{2}\int dE(-f'(E))[g^{(0)}_{\alpha\beta}(E)-ie\omega g^{(1)}_{\alpha\beta}(E)], \tag{29}$$

where $g^{(0)}_{\alpha\beta}(E)=\mathcal{A}_{\alpha\beta}(E,0)$ is the linear dc conductance, and

$$g^{(1)}_{\alpha\beta}(E)=\int d\mathbf{r}\left[\frac{dn_{\alpha\beta}(E,\mathbf{r})}{dE}-u_\beta(E,\mathbf{r})\frac{dn_\alpha(E,\mathbf{r})}{dE}\right] \tag{30}$$

is the linear emittance corresponding to the frequency-dependent part of the linear conductance, which has been given by Büttiker.[16] The relations $dN_{\alpha\beta}(E)/dE=\int d\mathbf{r}dn_{\alpha\beta}(E,\mathbf{r})/dE$ and

$$\frac{dn_{\alpha\beta}(E,\mathbf{r})}{dE}=-\frac{1}{4\pi i}\mathrm{Tr}\left[S^\dagger_{\alpha\beta}(E)\frac{dS_{\alpha\beta}(E)}{dU(\mathbf{r})}-\frac{dS^\dagger_{\alpha\beta}(E)}{dU(\mathbf{r})}S_{\alpha\beta}(E)\right] \tag{31}$$

have been used in the derivation of Eq. (30). $dn_{\alpha\beta}(E,\mathbf{r})/dE$ is the partial LDOS and $dn_\alpha(E,\mathbf{r})/dE=\Sigma_\beta dn_{\alpha\beta}(E,\mathbf{r})/dE$ is the LDOS. The trace in Eq. (31) is taken over the channel index k. We will suppress the trace in the following discussions but keep it in mind that there is a trace which should take over the channel index.

The first-order nonlinear conductance is given by

$$G_{\alpha\beta\gamma}(\omega)=\delta(\omega)G^{(0)}_{\alpha\beta\gamma}+[\delta(\omega+2\Omega)+\delta(\omega-2\Omega)]G^{(2\Omega)}_{\alpha\beta\gamma}(\omega), \tag{32}$$

with $G^{(0)}_{\alpha\beta\gamma}=2G^{(2\Omega)}_{\alpha\beta\gamma}(0)$ and

$$G^{(2\Omega)}_{\alpha\beta\gamma}(\omega)=\frac{e^3}{2^3h}\int dE(-f'(E))\left[g^{(0)}_{\alpha\beta\gamma}(E)-ie\omega\int dE(-f'(E))g^{(1)}_{\alpha\beta\gamma}(E)\right], \tag{33}$$

where

$$g^{(0)}_{\alpha\beta\gamma}(E)=\delta_{\beta\gamma}\partial_E g^{(0)}_{\alpha\beta}(E)+\int d\mathbf{r}\left[\left(u_\gamma(E,\mathbf{r})\partial_U-\frac{du_\gamma(E,\mathbf{r})}{dE}\right)g^{(0)}_{\alpha\beta}(E)\right]+[\beta\leftrightarrow\gamma)]\} \tag{34}$$

and

$$g^{(1)}_{\alpha\beta\gamma}(E)=-\int d\mathbf{r}\left\{u_{\beta\gamma}(E,\mathbf{r})\frac{dn_\alpha(E,\mathbf{r})}{dE}-\delta_{\beta\gamma}\frac{d^2n_{\alpha\beta}(E,\mathbf{r})}{dE^2}-\int d\mathbf{r}_1 u_\beta(E,\mathbf{r})u_\gamma(E,\mathbf{r}_1)\frac{d^2n_\alpha(E,\mathbf{r})}{dE^2}+\left[u_\gamma(E,\mathbf{r})\frac{d^2n_{\alpha\beta}(E,\mathbf{r})}{dE^2}+g_{\alpha\beta}(E)\times\int d\mathbf{r}_1\frac{du_\gamma(E,\mathbf{r}_1)}{dE}+\frac{1}{4\pi i}\left(\frac{du_\gamma(E,\mathbf{r})}{dE}\partial_U-\frac{d^2u_\gamma(E,\mathbf{r})}{dE^2}\right)g^{(0)}_{\alpha\beta}(E)+(\beta\leftrightarrow\gamma)\right]\right\}. \tag{35}$$

From Eqs. (34) and (35), it is found that the internal interaction contributes itself to both dc and ac aspects in the nonlinear response. The same thing happens in the third-order conductance too. The displacement current contributes itself to dc and ac components of the nonlinear current in the response of the voltage.

In the same way, the third-order conductance is obtained as

$$G_{\alpha\beta\gamma\rho}(\omega)=[\delta(\omega+\Omega)+\delta(\omega-\Omega)]G^{(\Omega)}_{\alpha\beta\gamma\rho}(\omega)+[\delta(\omega+3\Omega)+\delta(\omega-3\Omega)]G^{(3\Omega)}_{\alpha\beta\gamma\rho}(\omega), \tag{36}$$

with

$$G^{(\Omega)}_{\alpha\beta\gamma\rho}(\omega)=3G^{(\Omega)}_{\alpha\beta\gamma\rho}(\omega)=\frac{e^4}{2^4h}\int dE(-f'(E))[g^{(0)}_{\alpha\beta\gamma\rho}(E)-ie\omega g^{(1)}_{\alpha\beta\gamma\rho}(E)], \tag{37}$$

where

$$g^{(0)}_{\alpha\beta\gamma\rho}(E)=\delta_{\beta\gamma}\delta_{\beta\rho}\partial^2_E g^{(0)}_{\alpha\beta}(E)+\int d\mathbf{r}\left\{\left[u_{\gamma\rho}(E,\mathbf{r})\partial_U+\delta_{\beta\gamma}u_\rho(E,\mathbf{r})\partial_E\partial_U+\int d\mathbf{r}_1 u_\gamma(E,\mathbf{r})u_\rho(E,\mathbf{r}_1)\partial^2_U\right]g^{(0)}_{\alpha\beta}(E)\right\}_c-\int d\mathbf{r}\left\{\left[\frac{du_{\gamma\rho}(E,\mathbf{r})}{dE}+\left(\delta_{\beta\gamma}\frac{d^2u_\rho(E,\mathbf{r})}{dE^2}-\int d\mathbf{r}_1\frac{d^2}{dE^2}[u_\gamma(E,\mathbf{r})u_\rho(E,\mathbf{r}_1)]\right)-2\left(\delta_{\beta\gamma}\frac{du_\rho(E,\mathbf{r})}{dE}\partial_U A_{\beta\beta}(\alpha,E,E)-\int d\mathbf{r}_1\frac{d}{dE}[u_\gamma(E,\mathbf{r})u_\rho(E,\mathbf{r}_1)]\partial_U\right)\right]g^{(0)}_{\alpha\beta}(E)\right\}_c \tag{38}$$

and

$$
\begin{aligned}
g^{(1)}_{\alpha\beta\gamma\rho}(E) = & -\int d\mathbf{r}\Bigg\{ u_{\beta\gamma\rho}(E,\mathbf{r})\frac{dn_\alpha(E,\mathbf{r})}{dE} + \int d\mathbf{r}_1 \int d\mathbf{r}_2 u_\beta(E,\mathbf{r}) u_\gamma(E,\mathbf{r}_1) u_\rho(E,\mathbf{r}_2) \\
& \times \partial_U^2 \frac{dn_\alpha(E,\mathbf{r})}{dE} - \delta_{\beta\gamma}\delta_{\beta\rho}\frac{d^3 n_{\alpha\beta}(E,\mathbf{r})}{dE^3} + \Bigg[u_{\gamma\rho}(E,\mathbf{r})\frac{d^2 n_{\alpha\beta}(E,\mathbf{r})}{dE^2} \\
& + \int d\mathbf{r}_1 u_\beta(E,\mathbf{r}_1) u_{\gamma\rho}(E,\mathbf{r})\partial_U \frac{dn_\alpha(E,\mathbf{r})}{dE} + \delta_{\beta\gamma} u_\rho(E,\mathbf{r})\frac{d^3 n_{\alpha\beta}(E,\mathbf{r})}{dE^3} \\
& + \int d\mathbf{r}_1 u_\gamma(E,\mathbf{r}) u_\rho(E,\mathbf{r}_1)\partial_U \frac{d^2 n_{\alpha\beta}(E,\mathbf{r})}{dE^2} + \delta_{\gamma\rho}\frac{d}{dE}\left(\frac{dn_{\alpha\gamma}(E,\mathbf{r})}{dE}\,\frac{du_\beta(E,\mathbf{r})}{dE}\right) \\
& - \int d\mathbf{r}_1 u_\rho(E,\mathbf{r}_1)\frac{d}{dE}\left(\frac{dn_{\alpha\gamma}(E,\mathbf{r})}{dE}\,\frac{du_\beta(E,\mathbf{r})}{dE}\right) - \int d\mathbf{r}_1 u_\gamma(E,\mathbf{r}_1)\frac{d}{dE}\left(\frac{dn_{\alpha\rho}(E,\mathbf{r})}{dE}\,\frac{du_\beta(E,\mathbf{r})}{dE}\right) \\
& + \int d\mathbf{r}_1 \int d\mathbf{r}_2 u_\gamma(E,\mathbf{r}_1) u_\rho(E,\mathbf{r}_2)\frac{d}{dE}\left(\frac{dn_\alpha(E,\mathbf{r})}{dE}\,\frac{du_\beta(E,\mathbf{r})}{dE}\right)\Bigg]_c\Bigg\} \\
& + \int d\mathbf{r}\left\{ g^{(1)}_{\alpha\beta}(E)\frac{du_{\gamma\rho}(E,\mathbf{r})}{dE} + g^{(1)}_{\alpha\gamma\rho}(E)\frac{du_\beta(E,\mathbf{r})}{dE}\right\}_c \\
& + \frac{1}{4\pi i}\int d\mathbf{r}\Bigg\{\Bigg[\frac{d^2 u_{\gamma\rho}(E,\mathbf{r})}{dE^2} - \frac{du_{\gamma\rho}(E,\mathbf{r})}{dE}\partial_U + \delta_{\beta\gamma}\left(\frac{d^3 u_\rho(E,\mathbf{r})}{dE^3} - 2\frac{d^2 u_\rho(E,\mathbf{r})}{dE^2}\partial_U - \frac{du_\rho(E,\mathbf{r})}{dE}\partial_U\partial_E\right) \\
& - \int d\mathbf{r}_1 u_\gamma(E,\mathbf{r}_1)\left(\frac{d^3 u_\rho(E,\mathbf{r})}{dE^3} - 2\frac{d^2 u_\rho(E,\mathbf{r})}{dE^2}\partial_U + \frac{du_\rho(E,\mathbf{r})}{dE}\partial_U^2\right) \\
& - \int d\mathbf{r}_1 u_\rho(E,\mathbf{r}_1)\left(\frac{d^3 u_\gamma(E,\mathbf{r})}{dE^3} - 2\frac{d^2 u_\gamma(E,\mathbf{r})}{dE^2}\partial_U + \frac{du_\gamma(E,\mathbf{r})}{dE}\partial_U^2\right) \\
& - \int d\mathbf{r}_1\left(4\frac{du_\gamma(E,\mathbf{r})}{dE}\,\frac{d^2 u_\rho(E,\mathbf{r}_1)}{dE^2} - 2\frac{du_\rho(E,\mathbf{r})}{dE}\,\frac{du_\gamma(E,\mathbf{r}_1)}{dE} + \frac{du_\gamma(E,\mathbf{r})}{dE}u_\rho(E,\mathbf{r}_1)\right) - (\gamma\leftrightarrow\rho)\Bigg] g^{(0)}_{\alpha\beta}(E)\Bigg\}_c .
\end{aligned}
\tag{39}
$$

In the view of Eqs. (29), (33), and (37), the conductances can be computed through the evaluation of quantities called the sensitivity $\mathcal{A}_{\alpha\beta}(E,0)$, the characteristic potential tensors, and the LDOS.

Under the Thomas-Fermi approximation, the current conservation and gauge invariance can be explicitly confirmed, i.e., the following sum rules are satisfied: $\Sigma_\alpha G_{\alpha\beta}(\omega) = \Sigma_\alpha G_{\alpha\beta\gamma}(\omega) = \Sigma_\alpha G_{\alpha\beta\gamma\rho}(\omega) = 0$, which means the current conservation, $\Sigma_\beta G_{\alpha\beta}(\omega) = \Sigma_\beta G_{\alpha\beta\gamma}(\omega) = \Sigma_\beta G_{\alpha\beta\gamma\rho}(\omega) = 0$ and $\Sigma_\gamma G_{\alpha\beta\gamma}(\omega) = \Sigma_\gamma G_{\alpha\beta\gamma\rho}(\omega) = \Sigma_\rho G_{\alpha\beta\gamma\rho}(\omega) = 0$, guarantee the gauge invariance of the theory.

III. APPLICATION

As examples, we apply the formulas of conductances in the last section to the geometrically asymmetric and symmetric systems of double-barrier tunneling diodes. Consider a one-dimensional double-barrier tunneling system where two barriers are δ functions located at positions $x=-a$ and $x=a$. The scattering matrix close to a resonance is given by the Breit-Wigner formula

$$
S_{\alpha\beta}(E) = \left[\delta_{\alpha\beta} - i\frac{\sqrt{\Gamma_\alpha\Gamma_\beta}}{E-E_r+i(\Gamma/2)}\right] e^{i(\phi_\alpha+\phi_\beta)}, \tag{40}
$$

where Γ_α is the partial width of resonance proportional to the tunneling probability through the probe α and $\Gamma = \Sigma_\alpha \Gamma_\alpha$ is the total width of resonance. ϕ_α are the phases acquired in the reflection or transmission process. Through some straightforward algebra and Eq. (31), the partial LDOS are found:

$$
\frac{dN_{\alpha\alpha}}{dE} = \frac{1}{2\pi}\frac{\Gamma_\alpha}{|\Delta|^2}\frac{(E-E_r)^2-(\Gamma/2)^2+\Gamma_\alpha(\Gamma/2)}{|\Delta|^2} \tag{41}
$$

and

$$
\frac{dN_{\alpha\beta}}{dE} = \frac{1}{2\pi}\frac{\Gamma_\alpha}{|\Delta|^2}\frac{\Gamma_\alpha(\Gamma/2)}{|\Delta|^2}, \quad \alpha\neq\beta, \tag{42}
$$

with $\Delta = (E-E_r) + i(\Gamma/2)$. The injectivities are

$$\frac{dN_\alpha}{dE}=\sum_\beta \frac{dN_{\alpha\beta}}{dE}=\frac{1}{2\pi}\frac{\Gamma_\alpha}{|\triangle|^2}. \tag{43}$$

From Eq. (43), the derivatives of injectivities with respect to the energy are then obtained:

$$\frac{d^2N_\alpha}{dE^2}=-\frac{1}{\pi}\frac{\Gamma_\alpha}{(|\triangle|^2)^2}(E-E_r) \tag{44}$$

and

$$\frac{d^3N_\alpha}{dE^3}=\frac{1}{\pi}\frac{\Gamma_\alpha}{(|\triangle|^2)^3}(3|\triangle|^2-\Gamma^2). \tag{45}$$

With the help of Eq. (41), the following derivatives of the LDOS with respect to the energy are found:

$$\frac{d^2N_{11}}{dE^2}=\frac{1}{\pi}\frac{\Gamma_1}{|\Delta|^2}\frac{\Gamma_2\Gamma-|\triangle|^2}{|\Delta|^2}\frac{E-E_r}{|\triangle|^2} \tag{46}$$

and

$$\frac{d^3N_{11}}{dE^3}=\frac{1}{\pi}\frac{\Gamma_1}{(|\triangle|^2)^4}\left[3(|\triangle|^2)^2-5|\triangle|^2\Gamma_2\Gamma -|\triangle|^2\Gamma^2+6\Gamma_2\Gamma\left(\frac{\Gamma}{2}\right)^2\right]. \tag{47}$$

In this example,the characteristic potential tensors are independent of $\mathbf{r}$ because of the Breit-Wigner formula for the scattering matrix. From Eqs. (B4)–(B6), the terms $\nabla^2 u_{\alpha\cdots}$ vanish under the Thomas-Fermi approximation. Then the characteristic potential tensors can be solved from Eqs. (B4)–(B6), evidently. Therefore, the characteristic potential tensors and their derivatives are obtained as

$$u_1=\frac{\Gamma_1}{\Gamma},\quad \frac{du_1}{dE}=0, \tag{48}$$

$$u_{11}=-2\frac{1}{|\triangle|^2}\frac{\Gamma_1\Gamma_2}{\Gamma^2}(E-E_r),$$

$$\frac{du_{11}}{dE}=\frac{1}{|\triangle|^2}\frac{\Gamma_1\Gamma_2}{\Gamma^2}\frac{2|\triangle|^2-\Gamma^2}{|\triangle|^2}, \tag{49}$$

and

$$\begin{aligned}u_{111}=&\left[\frac{dn}{dE}\right]^{-1}\left[\frac{d^3n_1}{dE}(1+3u^2-3u_1)-u_1^3\frac{d^3n}{dE^3}\right.\\&-3u_{11}\frac{d^3n_1}{dE^3}+3u_1u_{11}\frac{d^3n}{dE^3}+3(1-2u_1)\\&\left.\times\frac{d}{dE}\left(\frac{du_1}{dE}\frac{dn_1}{dE}\right)+3u_1^2\frac{d}{dE}\left(\frac{du_1}{dE}\frac{dn}{dE}\right)\right]\\=&-2\frac{1}{|\triangle|^2}\frac{3|\triangle|^2-\Gamma^2}{\Gamma^2}\frac{\Gamma_1\Gamma_2}{|\triangle|^2}\frac{\Gamma_1-\Gamma_2}{\Gamma}.\end{aligned} \tag{50}$$

Substituting these quantities into the expressions of conductances in Eqs. (29) and (30), the dc and ac linear conductances are obtained:

$$g_{11}^{(0)}(E)=\mathcal{T}(E)=\frac{\Gamma_1\Gamma_2}{|\triangle|^2} \tag{51}$$

and

$$g_{11}^{(1)}(E)=\frac{1}{2\pi}\frac{1}{\Gamma}\frac{|\triangle|^2-2(\Gamma/2)^2}{|\triangle|^2}\mathcal{T}(E), \tag{52}$$

where $\mathcal{T}(E)$ is the transmission coefficient.

By evaluating Eqs. (34) and (35) the first-order nonlinear conductances can be obtained as

$$g_{111}^{(0)}(E)=-\frac{\Gamma_1-\Gamma_2}{\Gamma}\frac{d\mathcal{T}(E)}{dE} \tag{53}$$

and

$$g_{111}^{(1)}(E)=-\frac{1}{2\pi}\frac{1}{|\triangle|^2}\frac{\Gamma_1-\Gamma_2}{\Gamma^2}(|\triangle|^2-\Gamma^2)\frac{d\mathcal{T}(E)}{E}. \tag{54}$$

It is found that because of the term

$$d\mathcal{T}(E)/dE=2(E-E_r)[(\Gamma_1-\Gamma_2)/\Gamma](\Gamma_1\Gamma_2/\Gamma),$$

G_{111} changes sign across the resonant point and hence can be negative.

In the same way, we can evidently calculate the second-order nonlinear conductances from Eqs. (38) and (39). It is found that

$$g_{1111}^{(0)}(E)=\frac{\Gamma_1^2+\Gamma_2^2-4\Gamma_1\Gamma_2}{\Gamma^2}\frac{d^2\mathcal{T}(E)}{dE^2} \tag{55}$$

and

$$\begin{aligned}g_{1111}^{(1)}(E)=&-\left\{u_{111}\frac{dN_1}{dE}-\frac{d^3N_{11}}{dE}+u_1^3\frac{d^3N_1}{dE^3}\right.\\&+3\left[u_{11}\frac{d^2N_{11}}{dE^2}-u_1u_{11}\frac{d^2N_1}{dE^2}+u_1\frac{d^3N_{11}}{dE^3}\right.\\&\left.-u_1^2\frac{d^3N_{11}}{dE^3}\right]-3g_{11}^{(1)}\frac{du_{11}}{dE}+\frac{3}{4\pi i}\\&\left.\times\left[\frac{d^2u_{11}}{dE^2}A_{11}(1,E,E)+\frac{du_{11}}{dE}\partial_E A_{11}(1,E,E)\right]\right\}\\=&\frac{3}{2\pi}\frac{1}{(|\triangle|^2)^4}\frac{\Gamma_1\Gamma_2}{\Gamma^3}\left[2(|\triangle|^2)^2(\Gamma_1^2+\Gamma_2^2-4\Gamma_1\Gamma_2)\right.\\&-|\triangle|^2\Gamma^2(4\Gamma_1^2+4\Gamma_2^2-11\Gamma_1\Gamma_2)+\Gamma^4\left(\Gamma_1^2+\Gamma_2^2\right.\\&\left.\left.-\frac{5}{2}\Gamma_1\Gamma_2\right)-i\Gamma_1\Gamma_2\Gamma(4|\triangle|^2-3\Gamma^2)(E-E_r)\right].\end{aligned} \tag{56}$$

All the components of conductances (linear and nonlinear) can be calculated in the same way. However, from the charge conservation and gauge invariance we have

$$G_{11}=-G_{12}=-G_{21}=G_{22}, \tag{57}$$

$$G_{111}=-G_{112}=-G_{121}=G_{122}=-G_{211} \\ =G_{212}=G_{221}=-G_{222}, \quad (58)$$

and

$$G_{1111}=-G_{2111}=-G_{1211}=-G_{1121}=-G_{1112}=G_{2112} \\ =G_{2121}=G_{2211}=G_{1212}=G_{1221}=G_{1122}=-G_{2122} \\ =-G_{2212}=-G_{2221}=-G_{1222}=G_{2222}. \quad (59)$$

For a geometrically symmetric system of double barriers, i.e., $\Gamma_1=\Gamma_2=\Gamma/2$, the dc and ac parts of linear and nonlinear conductances are obtained in the following forms:

$$g_{11}^{(0)}(E)=\mathcal{T}(E)=\frac{1}{4}\frac{\Gamma^2}{|\triangle|^2},$$

$$g_{11}^{(1)}(E)=\frac{1}{4\pi}\frac{1}{\Gamma}\frac{2|\triangle|^2-\Gamma^2}{|\triangle|^2}\mathcal{T}(E) \quad (60)$$

and

$$g_{1111}^{(0)}(E)=-\frac{1}{2}\frac{d\mathcal{T}(E)}{dE},$$

$$g_{1111}^{(1)}(E)=-\frac{3}{2^6\pi}\frac{\Gamma}{(|\triangle|^2)^4}[8(|\triangle|^2)^2-6|\triangle|^2\Gamma^2+\Gamma^4 \\ +i(4|\triangle|^2-3\Gamma^2)\Gamma(\triangle+\triangle^*)]. \quad (61)$$

The first-order nonlinear conductances vanish, i.e., $g_{111}^{(0)}(E)=g_{111}^{(1)}(E)=0$. The zero of the first-order nonlinear conductance $g_{111}(E)$ is evident.[19] In general, from $I_1=G_{11}(V_1-V_2)+G_{111}(V_1-V_2)^2+G_{1111}(V_1-V_2)^3+\cdots$ for a symmetric scattering volume with scattering potential $U(x)=U(-x)$ in one dimension where x is the propagation direction, we must have $-I_1$ if V_1 and V_2 are interchanged. Hence we conclude that for a symmetric scattering volume there are no quadratic terms, i.e., $G_{111}=0$. The first-order nonlinear conductance can be nonzero only for geometrically asymmetric systems. The sign of the nonlinear conductances can be positive or negative.

IV. SUMMARY

In this paper we have dealt with harmonic generation in the ac nonlinear response for a multiprobe mesoscopic system. The calculations are perturbatively carried up to third order at the low-frequency weakly nonlinear response of the probe voltages. By combining perturbation theory and scattering matrix theory, a model for the time-dependent system has been built. The theory has taken into account the oscillating applied potential and the oscillating internal self-induced potential nonlinearly. This allows us to obtain the response functions and the corresponding harmonic generation for not only their dc features but also those of the frequency dependence in a self-consistent way. The calculation proceeds by first solving the equation of motion and obtaining the solutions of a double operator in terms of the external voltage perturbatively. Then the current is written as an expansion in powers of voltage. The coefficients describing the total current flowing in and out of the system with respect to the probe voltages are functions of the unitary scattering matrix. As a main result, the harmonic generation has been shown step by step in our approach. Harmonic generation in the internal self-consistent potential is important for obtaining gauge-invariant and charge conservation results for each harmonic generation branch. How to separate out the higher harmonics for the nonlinear responses of the time-dependent external perturbation has been reported. As shown in Sec. II, in the generation of higher-frequency harmonics for the nonlinear components of conductances, there would be several branches at the same order of conductance. Each branch has its own characteristic frequency. For example, there is a branch of the dc part and one at twice the driving frequency for the transport current in the second-order response of the voltages. However, there is a branch with the oscillating frequency and a branch at triple the driving frequency for the transport current in the third-order response of voltages. The approach to higher-harmonic generation can been studied in the same framework built here. The conductances derived in this paper are expressed in terms of the Fermi surface quantities. Current conservation and gauge invariance are indeed satisfied by all Fourier component of the harmonics, which is consistent with physical requirements. It is found that the the internal interaction contributes to the dc aspect in the nonlinear regime. The displacement current contributes itself to the dc components of nonlinear current in the nonlinear response of voltage too. This is consistent with the conclusions of Refs. 21 and 22; the current in nonlinear regime is dependent on the band bottom position of the quantum well. We would like to emphasize that the magnetic field accompanied by the time-dependent internal potential is negligible in the weakly nonlinear response region because of a slow variation in time. However, the full Maxwell equations, namely, the Helmhotz equations describing both the internal self-induced potential and the corresponding vector potential, should be considered[5] if we consider the magnetic field induced by the time-dependent fluctuations of current. In the applications, the formulas of the ac conductances have been applied to a double-barrier tunneling diode using the Breit-Wigner form for the scattering matrix. The quadratic conductance would be absent for geometrically symmetric systems. The formulas of the dynamic conductances presented within this paper can be used in numerical simulations for any interesting sample because the formulas can be expressed in terms of the density of states explicitly.[19,23,24] Experimental verification of our approaches holds promise for a wide range of studies in the areas of quantum nonlinear transport.

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APPENDIX A: HAMILTONIAN AND ITS SOLUTIONS

The Hamiltonian density is

$$\hat{\mathcal{H}}=\hat{\mathcal{H}}_0+\hat{\mathcal{H}}_I=\frac{1}{2M}\left(\hat{\mathbf{P}}-\frac{e}{c}\mathbf{A}_0(\mathbf{r})\right)^2 +V_0(\mathbf{r})+eV(\mathbf{r},t)+\triangle E(\mathbf{r},\{V\},t), \tag{A1}$$

where $\mathbf{A}_0(\mathbf{r})$ is a steady vector potential, $\mathbf{B}=\nabla\times\mathbf{A}_0(\mathbf{r})$, and a $V_0(\mathbf{r})$ is a steady potential. $eV(\mathbf{r},t)$ is the perturbation Hamiltonian concerned with the external potential due to an application of voltages $V_\alpha e^{-\delta|t|}\cos\Omega t$ at the probes α. The term $\triangle E(\mathbf{r},\{V\},t)$ is the change in the energy which is the function of the internal potential $U(\mathbf{r},t)$ induced by a variation of the density of electrons after application of the external voltage.

The total Hamiltonian is then taken in the form

$$H=\int dEdE'\int d\mathbf{r}\Psi_E^\dagger(\mathbf{r})(\hat{\mathcal{H}}_0+\hat{\mathcal{H}}_I)\Psi_{E'}(\mathbf{r}), \tag{A2}$$

in which $\Psi_E(\mathbf{r})$ is the electron field operator. To make further progress, employing the matrix element of the current density,

$$\boldsymbol{\nabla}\cdot\mathbf{W}_{EE'}(\mathbf{r})=-\frac{2M}{\hbar^2}(E'-E)\Psi_E^\dagger(\mathbf{r})\Psi_{E'}(\mathbf{r}), \tag{A3}$$

with the notation $\mathbf{W}_{EE'}(\mathbf{r})=\Psi_E^\dagger(\mathbf{r})\hat{\mathbf{W}}(\mathbf{r})\Psi_{E'}(\mathbf{r})$, the Hamiltonian becomes

$$H=\int dEdE'\int d\mathbf{r}\bigg[\Psi_E^\dagger(\mathbf{r})\hat{\mathcal{H}}_0\Psi_{E'}(\mathbf{r}) -\frac{e\hbar^2}{2M}\frac{1}{E'-E}\boldsymbol{\nabla}\cdot\mathbf{W}_{EE'}(\mathbf{r})\tilde{V}(\mathbf{r},t)\bigg]. \tag{A4}$$

In order to write the Hamiltonian in the form of second quantization, we use the field operator

$$\Psi(\mathbf{r},t)=\sum_{\alpha m}\frac{1}{[hv_{\alpha m}(E_{\alpha m})]^{1/2}}\psi_{\alpha m}(E_{\alpha m},\mathbf{r})C_{\alpha m}(E_{\alpha m}) \times e^{-i(E_{\alpha m}/\hbar)t}, \tag{A5}$$

where $\psi_{\alpha m}(E_{\alpha m},\mathbf{r})$ is the wave function with a channel index m at the probe α, $C_{\alpha m}$ is the annihilation operator of electron in the incoming channel m inside the probe α, and $1/\hbar v_{\alpha m}(E)$ is the one-dimensional density of states of the quantum channel m at energy E.[2,16] Then the first term, i.e., the free electron Hamiltonian H_0, becomes

$$H_0=\int dE\sum_{\alpha m}EC_{\alpha m}^\dagger(E)C_{\alpha m}(E). \tag{A6}$$

The second term in Eq. (A4) is a perturbation Hamiltonian which can be derived by consideration of the boundary conditions that the applied voltages are constants at the perfect probes, the electric field is approximately unchanged in the finite region of space, and current conservation. Integrating the second term in Eq. (A4) and using the divergence theorem, it yields

$$\int d\mathbf{r}\boldsymbol{\nabla}\cdot\mathbf{W}_{EE'}(\mathbf{r})\tilde{V}(\mathbf{r})=i\frac{2M}{e\hbar}\sum_\alpha\tilde{V}_\alpha\int_{\mathcal{L}_\alpha}dy_\alpha\mathbf{J}_{EE'}(\mathbf{r})\cdot\hat{\mathbf{x}}_\alpha, \tag{A7}$$

where $\hat{\mathbf{x}}_\alpha$ is a unit vector parallel to the probe α and pointing outward from the junction region, $\mathcal{L}_\alpha$ is the cross-section curve of probe α, and $\int_{\mathcal{L}_\alpha}dy_\alpha$ denotes the integral over the cross section of it. Furthermore, it is found[2,16] that $\int_{\mathcal{L}_\alpha}dy_\alpha\mathbf{J}_{EE'}(\mathbf{r})\cdot\hat{\mathbf{x}}_\alpha=(e/h)\Sigma_m C_{\alpha m}^\dagger(E)C_{\alpha m}(E')$, which is available for the low-frequency situation. Hence, the corresponding Hamiltonian (A4) is rewritten in the form of second quantization,

$$H=\sum_{\alpha m}\int dEdE'\bigg[E\delta(E-E') +\frac{e}{2\pi i}\tilde{V}_\alpha(t)\frac{1}{E-E'}\bigg]C_{\alpha m}^\dagger(E,t)C_{\alpha m}(E',t), \tag{A8}$$

after considering the completeness and orthonormal conditions of the eigenstates of H_0, where $e\tilde{V}_\alpha(t)=eV_\alpha+\int d\mathbf{r}[\delta E_\alpha(U)/\delta U]U(\mathbf{r},t)$ and $eV_\alpha(t)$ is the shift of the electrochemical potential μ_α away from the equilibrium state associated with μ^{eq}. This is precisely the Hamiltonian of the charged particle system with the perturbation due to the presence of time-dependent external voltage at the probe α.

Now we present the perturbation solution of the double-operator $C_{\gamma k}^\dagger(E,t)C_{\gamma k}(E',t)$. Upon working to third order in powers of $\mathcal{B}_{\alpha\beta,mn}(E,E',t)$, the time dependences of the operators involved in the second term and in the third term in Eq. (8) are taken to those in the first-order perturbation system and the second-order perturbation system, respectively. The perturbation solution is obtained :

$$C_{\gamma k}^\dagger(E,t)C_{\gamma k}(E',t)=e^{-i(E'-E)t/\hbar}C_{\gamma k}^\dagger(E)C_{\gamma k}(E') +\hat{\mathcal{W}}_1(E,E',t)+\hat{\mathcal{W}}_2(E,E',t) +\hat{\mathcal{W}}_3(E,E',t), \tag{A9}$$

where three operators of $\hat{\mathcal{W}}_j(t)$ are given by

$$\hat{\mathcal{W}}_1(E,E',t)=\frac{1}{i\hbar}\frac{e}{2\pi i}e^{-i(E'-E)t/\hbar}\int_{-\infty}^{t}dt_1\sum_{\beta n}\int dE''\mathcal{B}_{\beta\gamma,nk}(E',E'',t_1)C^{\dagger}_{\gamma k}(E)C_{\beta n}(E'')e^{-i(E''-E')t_1/\hbar}$$
$$-\frac{1}{i\hbar}\frac{e}{2\pi i}e^{-i(E'-E)t/\hbar}\int_{-\infty}^{t}dt_1\sum_{\beta n}\int dE''\mathcal{B}_{\gamma\beta,kn}(E'',E,t_1)C^{\dagger}_{\beta n}(E'')C_{\gamma k}(E')e^{-i(E-E'')t_1/\hbar}, \quad \text{(A10)}$$

$$\hat{\mathcal{W}}_2(E,E',t)=\frac{1}{(i\hbar)^2}\left(\frac{e}{2\pi i}\right)^2\frac{1}{2!}e^{-i(E'-E)t/\hbar}\int dE''dE'''\sum_{\beta n,\delta m}\int_{-\infty}^{t}dt_1\mathcal{B}_{\beta\gamma,nk}(E',E'',t_1)e^{-i(E'-E'')t_1/\hbar}$$
$$\times\int_{-\infty}^{t_1}dt_2[\mathcal{B}_{\gamma\delta,kl}(E'',E''',t_2)C^{\dagger}_{\gamma k}(E)C_{\delta l}(E'')e^{-i(E'''-E'')t_2/\hbar}-\mathcal{B}_{\delta\gamma,lk}(E''',E,t_2)C^{\dagger}_{\delta l}(E''')C_{\gamma k}(E'')e^{-i(E-E''')t_2\hbar}]$$
$$-\frac{1}{(i\hbar)^2}\left(\frac{e}{2\pi i}\right)^2\frac{1}{2!}e^{-i(E'-E)t/\hbar}\int dE''dE'''\sum_{\beta n,\delta m}\int_{-\infty}^{t}dt_1\mathcal{B}_{\beta\gamma,nk}(E'',E',t_1)e^{-i(E'-E'')t_1/\hbar}$$
$$\times\int_{-\infty}^{t_1}dt_2[\mathcal{B}_{\gamma\delta,kl}(E',E''',t_2)C^{\dagger}_{\gamma k}(E'')C_{\delta l}(E''')e^{-i(E'''-E')t_2/\hbar}$$
$$-\mathcal{B}_{\delta\gamma,lk}(E''',E'',t_2)C^{\dagger}_{\delta l}(E''')C_{\gamma k}(E')e^{-i(E''-E''')t_2/\hbar}], \quad \text{(A11)}$$

and

$$\hat{\mathcal{W}}_3(E,E',t)=\left(\frac{1}{i\hbar}\right)^3\left(\frac{e}{2\pi i}\right)^3\frac{1}{3!}e^{-i(E'-E)t/\hbar}\sum_{\beta\gamma\rho,nml}\int dE''dE'''dE^{(4)}\int_{-\infty}^{t}dt_1\mathcal{B}_{\beta n,\alpha k}(E',E^{(4)},t_1)e^{-i(E^{(4)}-E')t_1/\hbar}$$
$$\times\int_{-\infty}^{t_1}dt_2\mathcal{B}_{\gamma m,\alpha k}(E^{(4)},E'',t_2)e^{-i(E''-E^{(4)})t_2/\hbar}\left[\int_{-\infty}^{t_2}dt_3\mathcal{B}_{\rho l,\alpha k}(E'',E''',t_3)e^{-i(E'''-E'')t_3/\hbar}C^{\dagger}_{\rho l}(E)C_{\rho l}(E''')\right.$$
$$\left.-\int_{-\infty}^{t_2}dt_3\mathcal{B}(E''',E,t_3)e^{-i(E-E''')t_3/\hbar}C^{\dagger}_{\rho l}(E''')C_{\rho l}(E'')\right]-\left(\frac{1}{i\hbar}\right)^3\left(\frac{e}{2\pi i}\right)^3\frac{1}{3!}e^{-i(E'-E)t/\hbar}$$
$$\times\sum_{\beta\gamma\rho,nml}\int dE''dE'''dE^{(4)}\int_{-\infty}^{t}dt_1\mathcal{B}_{\beta n,\alpha k}(E',E^{(4)},t_1)e^{-i(E^{(4)}-E')t_1/\hbar}\int_{-\infty}^{t_1}dt_2$$
$$\times\mathcal{B}_{\gamma m,\alpha k}(E'',E',t_2)e^{-i(E'-E'')t_2/\hbar}\left[\int_{-\infty}^{t_2}dt_3\mathcal{B}_{\rho l,\alpha k}(E^{(4)},E''',t_3)e^{-i(E'''-E^{(4)})t_3/\hbar}C^{\dagger}_{\rho l}(E'')C_{\rho l}(E''')\right.$$
$$\left.-\int_{-\infty}^{t_2}dt_3\mathcal{B}(E''',E'',t_3)e^{-i(E''-E''')t_3/\hbar}C^{\dagger}_{\rho l}(E''')C_{\rho l}(E^{(4)})\right]-\left(\frac{1}{i\hbar}\right)^3\left(\frac{e}{2\pi i}\right)^3\frac{1}{3!}e^{-i(E'-E)t/\hbar}$$
$$\times\sum_{\beta\gamma\rho,nml}\int dE''dE'''dE^{(4)}\int_{-\infty}^{t}dt_1\mathcal{B}_{\beta n,\alpha k}(E^{(4)},E,t_1)e^{-i(E-E^{(4)})t_1/\hbar}\int_{-\infty}^{t_1}dt_2$$
$$\times\mathcal{B}_{\gamma m,\alpha k}(E',E'',t_2)e^{-i(E''-E)t_2/\hbar}\left[\int_{-\infty}^{t_2}dt_3\mathcal{B}_{\rho l,\alpha k}(E'',E''',t_3)e^{-i(E'''-E'')t_3/\hbar}C^{\dagger}_{\rho l}(E^{(4)})C_{\rho l}(E''')\right.$$
$$\left.-\int_{-\infty}^{t_2}dt_3\mathcal{B}_{\rho l,\alpha k}(E''',E^{(4)},t_3)e^{-i(E^{(4)}-E''')t_3/\hbar}C^{\dagger}_{\rho l}(E''')C_{\rho l}(E'')\right]+\left(\frac{1}{i\hbar}\right)^3\left(\frac{e}{2\pi i}\right)^3\frac{1}{3!}e^{-i(E'-E)t/\hbar}$$
$$\times\sum_{\beta\gamma\rho,nml}\int dE''dE'''dE^{(4)}\int_{-\infty}^{t}dt_1\mathcal{B}_{\beta n,\alpha k}(E^{(4)},E,t_1)e^{-i(E-E^{(4)}/\hbar)t_1}\int_{-\infty}^{t_1}dt_2$$
$$\times\mathcal{B}_{\gamma m,\alpha k}(E'',E^{(4)},t_2)e^{-i(E^{(4)}-E'')t_2/\hbar}\left[\int_{-\infty}^{t_2}dt_3\mathcal{B}_{\rho l,\alpha k}(E',E''',t_3)e^{-i(E'''-E')t_3/\hbar}C^{\dagger}_{\rho l}(E'')C_{\rho l}(E''')\right.$$
$$\left.-\int_{-\infty}^{t_2}dt_3\mathcal{B}_{\rho l,\alpha k}(E''',E'',t_3)e^{-i(E''-E''')t_3/\hbar}C^{\dagger}_{\rho l}(E'')C_{\rho l}(E')\right]. \quad \text{(A12)}$$

Here the intrinsic permutation-symmetry property has been considered because we are concerned with calculating the response functions. The response functions are invariant under interchange of the pairs β and γ for the second-order perturbation terms, and are invariant under permutations of the indices β, γ, and ρ for the third-order perturbation terms. This property, and its generalization to higher orders, is known as the ''intrinsic permutation symmetry.'' In the following calculations the intrinsic permutation-symmetry property indicates that the response functions (conductances, capacitances, and characteristic potential tensors) are invariant under any of the $n!$ permutations of the n triplets $(\alpha,\omega,\mathbf{r})$.

To calculate the electric current we need the quantum statistical average of the double operator. With the help of $\langle C^{\dagger}_{\alpha k}(E) C_{\beta k}(E')\rangle_{eq} = \delta_{\alpha\beta}\delta(E-E')f_{\alpha}$, Eq. (17) has been obtained. The last three terms $\mathcal{W}_j(t)$ in Eq. (17) are given by the following expressions:

$$\mathcal{W}_1(t) = \frac{1}{i\hbar}\frac{e}{2\pi i} e^{-i(E'-E)t/\hbar}\int_{-\infty}^{t} dt_1 \sum_{\beta n}\int dE'' \frac{1}{E'-E''}\delta_{\beta\alpha}\delta_{nk} f_{\beta}(E'')\,\delta(E-E'')\tilde{V}_{\beta}(t_1) e^{-i(E''-E')t_1/\hbar}$$
$$-\frac{1}{i\hbar}\frac{e}{2\pi i} e^{-i/(E'-E'')t/\hbar}\int_{-\infty}^{t} dt_1 \frac{e}{2}\sum_{\beta n}\int dE'' \frac{1}{E''-E}\delta_{\beta\alpha}\delta_{nk} f_{\beta}(E'')\,\delta(E''-E')\tilde{V}_{\beta}(t_1) e^{-i(E-E'')t_1/\hbar}, \tag{A13}$$

$$\mathcal{W}_2(t) = \frac{1}{(i\hbar)^2}\left(\frac{e}{2\pi i}\right)^2 \frac{1}{2!} e^{-i(E'-E)t/\hbar}\int dE'' dE''' \sum_{\beta n,\gamma m}\int_{-\infty}^{t} dt_1 \frac{1}{E'-E''}\tilde{V}_{\beta}(t_1)\,\delta_{\beta\alpha}\delta_{nk} e^{-i(E'-E'')t_1/\hbar}$$
$$\times\int_{-\infty}^{t_1} dt_2\left[\frac{1}{E''-E'''}\tilde{V}_{\gamma}(t_2)\,\delta_{\gamma\beta}\delta_{mn} f_{\gamma}(E'')\,\delta(E-E'') e^{-i(E'''-E'')t_2/\hbar}\right.$$
$$\left.-\frac{1}{E'''-E}\tilde{V}_{\gamma}(t_2)\,\delta_{\gamma\beta}\delta_{mn} f_{\gamma}(E'')\,\delta(E'''-E'') e^{-i(E-E''')t_2/\hbar}\right]$$
$$-\frac{1}{(i\hbar)^2}\left(\frac{e}{2\pi i}\right)^2 \frac{1}{2!} e^{-i(E'-E)t/\hbar}\int dE'' dE''' \sum_{\beta n,\gamma m}\int_{-\infty}^{t} dt_1 \frac{1}{E''-E'}\tilde{V}_{\beta}(t_1)\,\delta_{\beta\alpha}\delta_{nk} e^{-i(E'-E'')t_1/\hbar}$$
$$\times\int_{-\infty}^{t_1} dt_2\left[\frac{1}{E'-E'''}\tilde{V}_{\gamma}(t_2)\,\delta_{\gamma\beta}\delta_{mn} f_{\gamma}(E'')\,\delta(E''-E''') e^{-i(E'''-E')t_2/\hbar}\right.$$
$$\left.-\frac{1}{E'''-E''}\tilde{V}_{\gamma}(t_2)\,\delta_{\gamma\beta}\delta_{mn} f_{\gamma}(E''')\,\delta(E'''-E') e^{-i(E''-E''')t_2/\hbar}\right], \tag{A14}$$

and

$$\mathcal{W}_3(t) = \left(\frac{1}{i\hbar}\right)^3\left(\frac{e}{2\pi i}\right)^3 \frac{1}{3!} e^{-i(E'-E)t/\hbar}\sum_{\beta\gamma\rho,nml}\delta_{\beta\alpha}\delta_{nk}\delta_{\gamma\beta}\delta_{mn}\delta_{\rho\gamma}\delta_{lm}\int dE'' dE''' dE^{(4)}$$
$$\times\int_{-\infty}^{t} dt_1 \frac{1}{E'-E^{(4)}}\tilde{V}_{\beta}(t_1) e^{-i(E^{(4)}-E')t_1/\hbar}\int_{-\infty}^{t_1} dt_2 \frac{1}{E^{(4)}-E''}\tilde{V}_{\gamma}(t_2) e^{-i(E''-E^{(4)}/\hbar)t_2}$$
$$\times\left[\int_{-\infty}^{t_2} dt_3 \frac{1}{E''-E'''}\tilde{V}_{\rho}(t_3) e^{-i(E'''-E'')t_3/\hbar} f_{\rho}(E)\,\delta(E-E''')\right.$$
$$\left.-\int_{-\infty}^{t_2} dt_3 \frac{1}{E'''-E}\tilde{V}_{\rho}(t_3) e^{-i(E-E''')t_3/\hbar} f_{\rho}(E''')\,\delta(E'''-E'')\right]$$
$$-\left(\frac{1}{i\hbar}\right)^3\left(\frac{e}{2\pi i}\right)^3 \frac{1}{3!} e^{-i(E'-E)t/\hbar}\sum_{\beta\gamma\rho,nml}\delta_{\beta\alpha}\delta_{nk}\delta_{\gamma\beta}\delta_{mn}\delta_{\rho\gamma}\delta_{lm}\int dE'' dE''' dE^{(4)}$$
$$\times\int_{-\infty}^{t} dt_1 \frac{1}{E'-E^{(4)}}\tilde{V}_{\beta}(t_1) e^{-i(E^{(4)}-E')t_1/\hbar}\int_{-\infty}^{t_1} dt_2 \frac{1}{E''-E'}\tilde{V}_{\gamma}(t_2) e^{-i(E'-E'')t_2/\hbar}$$
$$\times\left[\int_{-\infty}^{t_2} dt_3 \frac{1}{E^{(4)}-E'''}\tilde{V}_{\rho}(t_3) e^{-i(E'''-E^{(4)})t_3/\hbar} f_{\rho}(E'')\,\delta(E''-E''')\right.$$
$$\left.-\int_{-\infty}^{t_2} dt_3 \frac{1}{E'''-E''}\tilde{V}_{\rho}(t_3) e^{-i(E''-E''')t_3/\hbar} f_{\rho}(E''')\,\delta(E'''-E^{(4)})\right]$$

$$-\left(\frac{1}{i\hbar}\right)^3\left(\frac{e}{2\pi i}\right)^3\frac{1}{3!}e^{-i(E'-E)t/\hbar}\sum_{\beta\gamma\rho,nml}\delta_{\beta\alpha}\delta_{nk}\delta_{\gamma\beta}\delta_{mn}\delta_{\rho\gamma}\delta_{lm}\int dE''dE'''dE^{(4)}$$
$$\times\int_{-\infty}^{t}dt_1\frac{1}{E^{(4)}-E}\tilde{V}_\beta(t_1)e^{-i(E-E^{(4)})t_1/\hbar}\int_{-\infty}^{t_1}dt_2\frac{1}{E'-E''}\tilde{V}_\gamma(t_2)e^{-i(E''-E)t_2/\hbar}$$
$$\times\left[\int_{-\infty}^{t_2}dt_3\frac{1}{E''-E'''}\tilde{V}_\rho(t_3)e^{-i(E'''-E'')t_3/\hbar}f_\rho(E^{(4)})\,\delta(E^{(4)}-E''')\right.$$
$$\left.-\int_{-\infty}^{t_2}dt_3\frac{1}{E'''-E^{(4)}}\tilde{V}_\rho(t_3)e^{-i(E^{(4)}-E''')t_3/\hbar}f_\rho(E''')\,\delta(E'''-E'')\right]$$
$$+\left(\frac{1}{i\hbar}\right)^3\left(\frac{e}{2\pi i}\right)^3\frac{1}{3!}e^{-i(E'-E)t/\hbar}\sum_{\beta\gamma\rho,nml}\delta_{\beta\alpha}\delta_{nk}\delta_{\gamma\beta}\delta_{mn}\delta_{\rho\gamma}\delta_{lm}\int dE''dE'''dE^{(4)}$$
$$\times\int_{-\infty}^{t}dt_1\frac{1}{E^{(4)}-E}\tilde{V}_\beta(t_1)e^{-i(E-E^{(4)})t_1/\hbar}\int_{-\infty}^{t_1}dt_2\frac{1}{E''-E^{(4)}}\tilde{V}_\gamma(t_2)e^{-i(E^{(4)}-E'')t_2/\hbar}$$
$$\times\left[\int_{-\infty}^{t_2}dt_3\frac{1}{E'-E'''}\tilde{V}_\rho(t_3)e^{-i(E'''-E')t_3/\hbar}f_\rho(E'')(E''-E''')\right.$$
$$\left.-\int_{-\infty}^{t_2}dt_3\frac{1}{E'''-E''}\tilde{V}_\rho(t_3)e^{-i(E''-E''')t_3/\hbar}f_\rho(E'')\,\delta(E''-E')\right]. \tag{A15}$$

$\tilde{V}_\beta(t)$ is the global voltage on the probes β, which is discussed in Appendix B.

APPENDIX B: SELF-CONSISTENCE AND THE INDUCED INTERNAL POTENTIAL

We now address the problem of accounting for the Coulomb interaction in calculations of the response function due to the external voltage. The application of the voltage $V_\alpha(t)$ at the probes induces a charge fluctuation in the average value of $\tilde{V}_\alpha$; i.e., the time-dependent probe voltage can create a variation in the electron density. Hence, the total perturbation to which the electron system responds is the sum of the applied perturbation and internal potentials due to the induced variation of the electron density. When we neglect those effects due to the induced magnetic field, which are practically very weak in the weakly frequency-dependent approach, the induced internal potential U in the sample is determined by Poisson's equation

$$\nabla^2U(\mathbf{r},t)=-4\pi e\,\delta n(\mathbf{r},t), \tag{B1}$$

where the induced variation of density is denoted by $\delta n(\mathbf{r},t)=\Sigma_\alpha\delta n_\alpha(\mathbf{r},t)$. Considering the harmonic generations of the driving frequency, the induced potential will have the same space-dependence and some integer times the frequency in the time dependence as the applied perturbation. In the response to the applied voltage and the induced self-consistent potential, there are two contributions to the electron density at the probe α: the injected charge density due to variation of the electrochemical potential and the induced charge density $\delta n_{ind,\alpha}(\mathbf{r},t)$ due to the internal potential, respectively. In the Thomas-Fermi approximation, the variation of electron density is obtained:

$$\delta n_\alpha(\mathbf{r},t)=\frac{dn_\alpha(\mathbf{r})}{dE}\left\{e[V_\alpha(t)-U(t)]+\left[e\frac{d}{dE}[V_\alpha(t)-U(t)]\right]\left[e[V_\alpha(t)-U(t)]\right.\right.$$
$$\left.\left.+\left(e\frac{d}{dE}(V_\alpha(t)-U(t)]\right)e[V_\alpha(t)-U(t)]\right]+\frac{1}{2}\left[e\frac{d^2}{dE^2}[V_\alpha(t)-U(t)]\right]e^2[V_\alpha(t)-U(t)]^2\right\}$$
$$+\frac{1}{2}\frac{d^2n_\alpha(\mathbf{r})}{dE^2}\left\{e[V_\alpha(t)-U(t)]+\left[e\frac{d}{dE}[V_\alpha(t)-U(t)]\right]e[V_\alpha(t)-U(t)]\right\}$$
$$\times\left\{e[V_\alpha(t)-U(t)]+\left[e\frac{d}{dE}[V_\alpha(t)-U(t)]\right]e[V_\alpha(t)-U(t)]\right\}$$
$$+\frac{1}{3!}\frac{d^3n_\alpha(\mathbf{r})}{dE^3}e^3[V_\alpha(t)-U(t)]^3+O((V_\alpha-U)^4). \tag{B2}$$

In the weakly nonlinear region, the induced self-consistent potential can be expanded in powers of variation of electrochemical potential $d\mu_\alpha(t)=eV_\alpha(t)$, i.e.,

$$U(\mathbf{r},t)=\sum_\alpha u_\alpha(\mathbf{r})V_\alpha(t)+\frac{1}{2}e\sum_{\alpha\beta} u_{\alpha\beta}(\mathbf{r})V_\alpha(t)V_\beta(t)+\frac{1}{3!}e^2\sum_{\alpha\beta\gamma} u_{\alpha\beta\gamma}(\mathbf{r})V_\alpha(t)V_\beta(t)V_\gamma(t)+O((V_\alpha)^4), \tag{B3}$$

in which the coefficients are the characteristic potential u_α and the second-order characteristic potential tensor $u_{\alpha\beta}$ (which is symmetric in α and β). $u_{\alpha\beta\gamma}$ and higher-order terms are third-order and higher-order characteristic potential tensors. There are several sum rules on the characteristic potential tensors. If all the changes in the electrochemical potentials are the same, i.e., $d\mu_\alpha=d\mu$ for arbitrary index α, this corresponds to an overall shift of the electrostatic potential $edU-d\mu$. It implies that $\Sigma_\beta \mu_\beta=1$ and $\Sigma_{\beta\gamma}u_{\beta\gamma}=\Sigma_{\beta\gamma\rho}u_{\beta\gamma\rho}=0$. Due to gauge invariance, it is further found that $\Sigma_\beta u_{\beta\gamma}=\Sigma_\gamma u_{\beta\gamma}=0$ and $\Sigma_\beta u_{\beta\gamma\rho}=\Sigma_\gamma u_{\beta\gamma\rho}=\Sigma_\rho u_{\beta\gamma\rho}=0$. The sum rules for the higher-order characteristic potential tensors can be derived in a similar way.

Substituting Eqs. (B2) and (B3) into Poisson's equation (B1), the following equations for the first three of characteristic potential tensors are arrived at:

$$-\nabla^2 u_\beta(\mathbf{r})+4\pi e\frac{dn(\mathbf{r})}{dE}u_\beta(\mathbf{r})=4\pi e\frac{dn_\beta(\mathbf{r})}{dE}, \tag{B4}$$

$$\begin{aligned}-\nabla^2 u_{\beta\gamma}(\mathbf{r})+4\pi e\frac{dn(\mathbf{r})}{dE}u_{\beta\gamma}(\mathbf{r})=4\pi e\Bigg\{&\delta_{\beta\gamma}\frac{d^2n_\beta(\mathbf{r})}{dE^2}+u_\beta(\mathbf{r})\int d\mathbf{r}_1 u_\gamma(\mathbf{r}_1)\frac{d^2n(\mathbf{r})}{dE^2}-\Bigg[u_\gamma(\mathbf{r})\frac{d^2n_\beta(\mathbf{r})}{dE^2}\\&+\left(\frac{dn_\beta(\mathbf{r})}{dE}-\int d\mathbf{r}_1 u_\beta(\mathbf{r}_1)\frac{dn(\mathbf{r})}{dE}\right)\frac{du_\gamma(\mathbf{r})}{dE}\Bigg]-[\beta\leftrightarrow\gamma]\Bigg\},\end{aligned} \tag{B5}$$

and

$$\begin{aligned}-\nabla^2 u_{\beta\gamma\rho}(\mathbf{r})+4\pi e\frac{dn(\mathbf{r})}{dE}u_{\beta\gamma\rho}(\mathbf{r})=4\pi e\Bigg(&\delta_{\beta\gamma}\delta_{\beta\rho}\frac{d^3n_\beta(\mathbf{r})}{dE^3}-u_\beta(\mathbf{r})\int d\mathbf{r}_1 u_\gamma(\mathbf{r}_1)\int d\mathbf{r}_2 u_\rho(\mathbf{r}_2)\frac{d^3n(\mathbf{r})}{dE^3}\\&-\Bigg[\delta_{\gamma\rho}u_\beta(\mathbf{r})\frac{d^3n_\gamma(\mathbf{r})}{dE^3}-u_\gamma(\mathbf{r})\int d\mathbf{r}_1 u_\rho(\mathbf{r}_1)\frac{d^3n_\beta(\mathbf{r})}{dE^3}+u_{\gamma\rho}(\mathbf{r})\frac{d^2n_\beta(\mathbf{r})}{dE^2}\\&-\int d\mathbf{r}_1 u_\beta(\mathbf{r}_1)u_{\gamma\rho}(\mathbf{r})\frac{d^2n(\mathbf{r})}{dE^2}\Bigg]_c+\Bigg[\delta_{\gamma\rho}\frac{d}{dE}\left(\frac{dn_\gamma(\mathbf{r})}{dE}\frac{du_\beta(\mathbf{r})}{dE}\right)\\&-\int d\mathbf{r}_1 u_\rho(\mathbf{r}_1)\frac{d}{dE}\left(\frac{dn_\gamma(\mathbf{r})}{dE}\frac{du_\beta(\mathbf{r})}{dE}\right)-\int d\mathbf{r}_1 u_\gamma(\mathbf{r}_1)\frac{d}{dE}\left(\frac{dn_\rho(\mathbf{r})}{dE}\frac{du_\beta(\mathbf{r})}{dE}\right)\\&+\int d\mathbf{r}_1 u_\gamma(\mathbf{r}_1)\int d\mathbf{r}_2 u_\rho(\mathbf{r}_2)\frac{d}{dE}\left(\frac{dn(\mathbf{r})}{dE}\frac{du_\beta(\mathbf{r})}{dE}\right)\Bigg]_c-\Bigg\{\Bigg[\int d\mathbf{r}_1 u_{\gamma\rho}(\mathbf{r}_1)\frac{dn(\mathbf{r})}{dE}\\&-\left(\delta_{\gamma\rho}\frac{d^2n_\gamma(\mathbf{r})}{dE^2}-\int d\mathbf{r}_1 u_\rho(\mathbf{r}_1)\frac{d^2n_\gamma(\mathbf{r})}{dE^2}-\int d\mathbf{r}_1 u_\gamma(\mathbf{r}_1)\frac{d^2n_\rho(\mathbf{r})}{dE^2}\right.\\&\left.+\int d\mathbf{r}_1 u_\gamma(\mathbf{r}_1)\int d\mathbf{r}_2 u_\rho(\mathbf{r}_2)\frac{d^2n(\mathbf{r})}{dE^2}\right)\Bigg]\frac{du_\beta(\mathbf{r})}{dE}-\left(\frac{dn_\beta(\mathbf{r})}{dE}-u_\beta(\mathbf{r})\frac{dn(\mathbf{r})}{dE}\right)\\&\times\int d\mathbf{r}_1\frac{du_{\gamma\rho}(\mathbf{r}_1)}{dE}-2\left(\frac{dn_\beta(\mathbf{r})}{dE}-u_\beta(\mathbf{r})\frac{dn(\mathbf{r})}{dE}\right)\int d\mathbf{r}_1\frac{du_\gamma(\mathbf{r}_1)}{dE}\int d\mathbf{r}_2\frac{du_\rho(\mathbf{r}_2)}{dE}\Bigg\}_c\Bigg),\end{aligned} \tag{B6}$$

where the subscript c stands for the cyclic permutation among the indices β, γ, and ρ in Eq. (B6). Note that there are ''intrinsic permutation symmetries'' for these equations; i.e., they are invariant under the permutation among these subscripts.

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