Higher harmonic generation in a mesoscopic conductor

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Harmonic generation is analyzed in the weakly ac nonlinear response by using scattering matrix theory. An explicit formulation of frequency-dependent conductances for harmonic generation has been developed. The theory includes contributions from the displacement current due to the internal interactions caused by the charge redistribution inside the conductor. There are several components that are oscillating at the harmonic frequencies in the nonlinear current. The frequency-dependent terms of real and imaginary parts in nonlinear conductances can be separated explicitly. The application of the formalism is demonstrated for a double-barrier nanostructure.

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The study of quantum transport in an electronic system has been treated as a scattering problem since Landauer's original work[1] over 20 years ago. A typical point of interest is the frequency-dependent current in response to an oscillating voltage. The ac linear and nonlinear responses in a multiprobe mesoscopic conductor have been studied[2–8] extensively in recent years. Experimentally, Taboryski *et al.*[9] reported that the nonlinear and asymmetric conductance oscillations have been observed in quantum point contacts with small bias voltages. Nonlinear phenomena in conducting materials such as photoinduced absorption, bleaching, and photoluminescence, etc., have received wide attention experimentally.[10] Theoretically, Büttiker and co-workers have developed a formalism for the dynamic conductances by taking into account contributions of the internal self-consistent potential.[4,5,11] According to electrodynamics, when a time-dependent external field is applied to a conductor, the charge distribution inside the conductor is driven away from its equilibrium pattern, which induces an internal potential inside the conductor. This internal potential opposes the changes of charge distribution and screens the external field. In their theory, another fundamental requirement is the gauge invariance, i.e., shifting the voltages in all leads by a constant value should not alter the results, thus the current depends only on the voltage differences. In ac transport, the current conservation and gauge invariance will not be satisfied unless the induced internal potential is properly taken into account.[4] The ac electric properties become more complicated due to the presence of the internal potential.

One interesting aspect in ac nonlinear transport is harmonic generation. The analogy with nonlinear optics[12] leads one to pursue harmonic generation in ac nonlinear transport. de Vegvar[13] has studied the second-harmonic transport response of multiprobe mesoscopic conductors at low frequency by using a perturbation theory, and he found that the second-harmonic current at low frequency is a non–Fermi-surface quantity. Recently, Pedersen and Büttiker[14] have pointed out that the ac nonlinear transport process, theoretically, should involve a dc component and the components at the frequencies of oscillation and its higher harmonics. However, these approaches only involved the frequency-independent part of nonlinear conductances; frequency-dependent conductances have not been included. Recent studies have shown the importance of contributions from the internal potential in higher-order nonlinear conductances.[15,16] Consequently, it motivates us to develop a formalism in which both the dc and the ac features of harmonic generation are treated self-consistently. The study of the ac nonlinear feature of harmonic generation in nanostructures might be of technological interest. In nonlinear optics, the development of lasers has brought about an increased interest in the efficiency of harmonic generation. Modern technology might also provide useful applications with harmonic generations in nanodevices.

The aim of this paper is to formulate harmonic generation in weakly ac nonlinear response using the scattering matrix method. There was an earlier attempt at studying harmonic generation by perturbation theory.[16] We found that the scattering matrix theory is much simpler and physical for studying ac nonlinear transport. We were able to arrive at the same results as obtained from the perturbation theory.[16] The generalized formulas of dynamic conductances are derived up to third-harmonic generation. The process exactly traces the contributions from the frequency-dependent internal potential. It is shown how the ac nonlinear feature of harmonic generation arises in electronic transport. It is found that there are several components oscillating at the harmonic frequencies in the nonlinear current. In the regime of nonlinear response, the self-consistent potential contributes not only to the ac components of the current but also to its dc components. The developed formalism contains both the dc feature and the frequency-dependent feature of conductance for each component of harmonic generation. The current conservation and gauge invariance are satisfied by all components of harmonic generation. The nonlinearity shows that, in addition to the major component oscillating at the input frequency Ω, the electric current contains significant components oscillating at higher harmonic frequencies 2Ω, 3Ω, . . . , as well as the dc component at zero frequency. This is analogous to the well-known harmonic distortion of signals in an electrical circuit, where the response is also nonlinear. In the regime of nonlinear response it is found that there are two parts in the frequency-dependent conductances, i.e., $\omega \,\mathrm{Re}\, G^{\mathrm{ac}}$ and $i\omega \,\mathrm{Im}\, G^{\mathrm{ac}}$. The real part $\mathrm{Re}\, G^{\mathrm{ac}}$ changes the total current in the regime of nonlinear response. The application of ac nonlinear response will be illustrated for a two-terminal nanostructure. The frequency-dependent quantum effect of $\omega\,\mathrm{Re}\,G^{\mathrm{ac}}$ is calculated in a geometrically symmetric double-barrier system. The formalism developed here will enable one to perform more realistic numerical simulations to study the properties of quantum transport in nanostructure devices.

The model considered here is a conductor attached to a number of probes that are extended as ideal leads to infinity. The voltage $V_\alpha(t)=V_\alpha h(t)$ at probe α is constant in space and varies with time as $h(t)=\cos\Omega t$. The voltage relates to the chemical potential of the reservoir through $\mu_\alpha=eV_\alpha$. The voltage at contact is only a well-defined quantity if the local electric fields vanish deep inside the contact. The effect of the magnetic part of the electromagnetic waves on the electrons is much weaker[7] and we neglect it in our discussion. The current operator has been derived by Büttiker,[3] $I_\alpha(t)=(e/\hbar)\Sigma_m\int dE\,dE'[\tilde{C}^{(\mathrm{in})+}_{\alpha m}(E)\tilde{C}^{(\mathrm{in})}_{\alpha m}(E')-\tilde{C}^{(\mathrm{out})+}_{\alpha m}(E)\,\tilde{C}^{(\mathrm{out})}_{\alpha m}(E')]\exp[i(E-E')t/\hbar]$, where $\tilde{C}^{(\mathrm{out})}_{\alpha m}(E)$ is the operator that annihilates a carrier in the outgoing channel m in probe α. The annihilation operator in the outgoing channel, $\tilde{C}^{(\mathrm{out})}_{\alpha m}$, is related to the annihilation operator in the incoming channel $\tilde{C}^{(\mathrm{in})}$ via the scattering matrix $S_{\alpha\beta mn}$, i.e., $\tilde{C}^{(\mathrm{out})}_{\alpha m}(E)=\Sigma_{\beta,n}S_{\alpha\beta mn}(E)\tilde{C}^{(\mathrm{in})}_{\alpha n}(E)$.[3,4] This formula is exact up to linear order of ω and for large frequency it is an approximation to a space-dependent expression of the current operator.[8] Then the current incident on the probe α is expressed[3] in the form

$$I_\alpha(t)=\frac{e}{h}\sum_{\beta,mn}\int dE\,dE'\,\tilde{C}^{+}_{\beta n}(E)A_{\beta\beta,nn}(\alpha,m,E,E')\times\tilde{C}_{\beta n}(E')\exp[i(E-E')t/\hbar], \tag{1}$$

where $A_{\beta\beta,nn}(\alpha,m,E,E')=\delta_{\alpha\beta}\delta_{mn}-S^{+}_{\alpha\beta mn}(E)S_{\alpha\beta mn}(E')$ is the current matrix. In Eq. (1), the upper index (in) has been suppressed for simplicity. The scattering matrix $S_{\alpha\beta,mn}$ has been used to describe the relationship between the incoming electron in channel n on the probe β and the outgoing electron in channel m on the probe α. The scattering matrix is a functional of energy and of internal self-consistent potential $U(\mathbf{r},\{V_\alpha(t)\})$. This internal potential is created by the variation of the density of electrons. The variation is induced by the time-dependent voltage on the probe. As a consequence of the time dependence of voltage, the redistribution of the charge, due to the variation of the density of electrons in the conductor, is also a function of time. It has Fourier components at the driving frequency of the external voltage and the frequencies of its harmonics. So the induced internal potential inside the conductor will oscillate at the frequencies of all harmonics. The induced internal potential U is determined by the Poisson equation $\nabla^2U(\mathbf{r},t)=-4\pi e\,\delta n(\mathbf{r},t)$, where $\delta n=\Sigma_\alpha\delta n_\alpha$ is the variation of the density of electrons. $e\,\delta n_\alpha$ is the variation of the density of charge injected into the volume of the conductor by the perturbation on the probe α.[4,11] There are two contributions in $e\,\delta n_\alpha$: the injected charge density due to the variation of chemical potential $d\mu_\alpha=eV_\alpha$ on the probe α, and the density of induced charge $e\,\delta n_{\mathrm{ind},\alpha}$ due to the internal potential. We have $\delta n_\alpha(\mathbf{r},t)=\Sigma_j(1/j!)[d^jn_\alpha(\mathbf{r},t)/dE^j][eV_\alpha(t)]^j+\delta n_{\mathrm{ind},\alpha}(\mathbf{r},t)$, where dn_α/dE is the injectivity and $d^jn_\alpha/dE^j(j>1)$ is the energy derivative of the injectivity. The density of the induced charge $e\,\delta n_{\mathrm{ind},\alpha}$ is a functional of external voltages and the internal self-consistent potential. In the regime of weakly nonlinear response, the internal potential can be expanded in a series of the variation of chemical potential $d\mu_\alpha$,[4,8] i.e., $U(\mathbf{r},t)=e\Sigma_\alpha u_\alpha(\mathbf{r})V_\alpha(t)+(e^2/2)\Sigma_{\alpha\beta}u_{\alpha\beta}(\mathbf{r})V_\alpha(t)V_\beta(t)+\cdots$, where eu_α is the characteristic potential, and $e^2u_{\alpha\beta}$ (which is symmetric in α and β) is the second-order characteristic potential tensor. There are several sum rules on these characteristic potential tensors. If the electrochemical potentials on all probes are changed by the same constant amount, i.e., $d\mu_\alpha=d\mu$ for the arbitrary index α, and the system ends at equilibrium, it corresponds to an overall shift of the electrostatic potential $edU-d\mu$.[4] It implies that $\Sigma_\alpha u_\alpha(\mathbf{r})=1$ and $\Sigma_\beta u_{\{\beta\gamma\cdots\}}(\mathbf{r})=0$, where Σ_β is the sum taken over any index among the indices $\{\beta\gamma\cdots\}$. With the help of the relations δn_α and U, we classify the terms oscillating at the same frequency in accordance with harmonics generation in both sides of the Poisson equation. The equations of characteristic potential tensors are obtained as

$$-\nabla^2u_{\{\beta\gamma\cdots\}}(\mathbf{r})+4\pi e\frac{dn}{dE}u_{\{\beta\gamma\cdots\}}=4\pi e\sum_\alpha\mathcal{F}_{\alpha\{\beta\gamma\cdots\}}(\mathbf{r}). \tag{2}$$

$u_{\{\beta\gamma\cdots\}}$ and $\mathcal{F}_{\alpha\{\beta\gamma\cdots\}}$ are invariant under the permutation of indices $\{\beta\gamma\cdots\}$. The first two of the $\mathcal{F}$'s are $\mathcal{F}_{\alpha\beta}=(dn_{\alpha\beta}/dE)$ and $\mathcal{F}_{\alpha\beta\gamma}=\delta_{\beta\gamma}(d\mathcal{F}_{\alpha\beta}/dE)+u_\beta u_\gamma\Sigma_\rho(d\mathcal{F}_{\alpha\rho}/dE)-[u_\gamma(d\mathcal{F}_{\alpha\beta}/dE)+(\mathcal{F}_{\alpha\beta}-u_\beta\Sigma_\rho\mathcal{F}_{\alpha\rho})\times(du_\gamma/dE)]-(\beta\leftrightarrow\gamma)$.

The electron Hamiltonian in the leads is given by the single-particle terms $H=\Sigma_{\alpha m}[E_{\alpha m}+eV_\alpha(t)]C^{+}_{\alpha m}C_{\alpha m}$, where $C_{\alpha m}(E_{\alpha m},t)$ is the annihilation operator for an electron in the incoming channel m on the probe α and $eV_\alpha(t)$ is the shift of chemical potential μ_α away from μ^{eq} in the equilibrium state. $E_{\alpha m}$ can be determined if the shape of the α lead is given. The internal interaction induces a fluctuation at the average value of V_α. By considering a series expansion of energy in powers of the potential landscape U, the external potential is replaced by the self-consistent potential: $E_{\alpha m}+eV_\alpha(t)\to E_{\alpha m}+e\tilde{V}_\alpha(t)$, where $\tilde{V}_\alpha$ is the global voltage on the probe α. Different from the external voltage, the internal potential comes from the charge accumulation in the region outside the leads. The response of charge to the time-dependent voltage causes the internal potential to contain high harmonic components. Thus, harmonic generation not only appears in the current-voltage characteristics but also in the induced internal potential-voltage characteristics. The redistribution of the charge in the system produces the internal potential, which modifies the voltage acting on the system. Therefore, the total perturbation to which the electron system responds is the sum of the applied perturbation and the induced internal potential due to the redistribution of charge. In the following, the channel subscript m is dropped for simplicity. The Fourier transform of global voltage is given by $\tilde{V}_\beta(\nu)=\Sigma_{j\gamma}V^{(j)}_\gamma(\beta)h_j(\nu)$, where the frequency-dependent factors $h_j(\nu)$ are the Fourier transformations of $h^j(t)$. $V_\gamma^{(1)}(\beta)=V_\gamma\{\delta_{\gamma\beta}+\int d\mathbf{r}\, u_\gamma(\mathbf{r})[\delta E_\beta(U)/\delta U(\mathbf{r})]\}$ and $V_\gamma^{(j)}(\beta)=(1/j!)V_{\gamma_1}\cdots V_{\gamma_j}\int d\mathbf{r}\, u_{\gamma_1\cdots\gamma_j}(\mathbf{r})[\delta E_\beta(U)/\delta U(\mathbf{r})]$ are the first- and jth-order ($j=2,3,\ldots,$) global voltages on the probe α, respectively. The C operator in Eq. (1) is determined through the dynamic equation by the lead Hamiltonian H given above. The operator $C_{\alpha m}(E_{\alpha m},t)$ satisfies the equation of motion $i\hbar\partial_t C_{\alpha m}(E_{\alpha m},t)=[C_{\alpha m}(E_{\alpha m},t),H]$, which can be integrated because the time dependence of H is simple. It is found that $C_{\alpha m}(E_{\alpha m},t)=C_{\alpha m}(E_{\alpha,m})\times\exp\{-i[E_{\alpha m}t+e\int dt\tilde{V}_\alpha(t)]/\hbar\}$,[4,8] where $\tilde{V}_\alpha(t)$ includes both the external voltage and internal potential. The solution can then be expressed in a series in powers of the voltage. Its Fourier transformation is given by

$$\begin{aligned}\tilde{C}_\alpha(E)&=\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\, C_\alpha(E_\alpha,t)e^{iEt/\hbar}\\ &=C_\alpha(\tilde{E})-\frac{1}{2\hbar\Omega}eV_\alpha^{(1)}C_\alpha^{(-)}(\tilde{E},\hbar\Omega)-\frac{1}{2^3\hbar\Omega}eV_\alpha^{(2)}C_\alpha^{(-)}(\tilde{E},2\hbar\Omega)+\frac{1}{2^3(\hbar\Omega)^2}(eV_\alpha^{(1)})^2[C_\alpha^{(+)}(\tilde{E},2\hbar\Omega)-2C_\alpha(\tilde{E})]\\ &\quad-\frac{1}{3!2^2\hbar\Omega}eV_\alpha^{(3)}[C_\alpha^{(-)}(\tilde{E},3\hbar\Omega)+9C_\alpha^{(-)}(\tilde{E},\hbar\Omega)]+\frac{1}{2^4(\hbar\Omega)^2}eV_\alpha^{(1)}eV_\alpha^{(2)}[C_\alpha^{(+)}(\tilde{E},3\hbar\Omega)-C_\alpha^{(+)}(\tilde{E},\hbar\Omega)]\\ &\quad-\frac{1}{3!2^3(\hbar\Omega)^3}(eV_\alpha^{(1)})^3[C_\alpha^{(-)}(\tilde{E},3\hbar\Omega)-3C_\alpha^{(-)}(\tilde{E},\hbar\Omega)],\end{aligned}\qquad(3)$$

where we have suppressed the index m so that C_α is the vector form of the operators $C_{\alpha m}$. In Eq. (3), $C_\alpha(\tilde{E})=C_\alpha[E-(1/2)e^2V_\alpha^{(2)}]$, which rises from the shift of variable, $C_\alpha^{(\pm)}(\tilde{E},j\hbar\Omega)=C_\alpha(\tilde{E}+j\hbar\Omega)\pm C_\alpha(\tilde{E}-j\hbar\Omega)$. From this solution, it is found that the time-dependent voltage leads to a multiplication. The physics in Eq. (3) is transparent: $C_\alpha(E\pm\hbar\omega)$ is just the one-photon sideband and $C_\alpha(E\pm2\hbar\omega)$ corresponds to the second-harmonic generation. Generally, in the energy representation the wave function becomes dispersed over all energies $E\pm j\hbar\Omega$ with an integer j. One of its signatures in the nonlinearity is the generation of current harmonics with frequencies higher than Ω. This means that the dependence of current on the ac bias is oscillating with harmonic frequencies of the driving frequency. This is caused by the quantum-mechanical interference of different components of the wave function, which is spread over a set of energies $E\pm j\hbar\Omega$. The current can be calculated in a straightforward way by substituting the solution in Eq. (3) into Eq. (1). In the evaluation of a quantum-statistical average, we assume that the modulation imposed on the system is so slow that the contacts can still be regarded as in a dynamic equilibrium state. Hence we have $\langle C_\alpha^+(E)C_\beta(E')\rangle=\delta_{\alpha\beta}\delta(E-E')f_\alpha(E)$, where $f_\alpha(E)$ is the Fermi function of reservoir α. The Fourier-transformed form of Eq. (1) is $I_\alpha(\omega)=(1/2\pi)\int_{-\infty}^{\infty}dt\, e^{i\omega t}I_\alpha(t)$. In order to show the nonlinear characteristics of the current in response to the external oscillating potential in the harmonic generation, we take the perturbation calculation up to the third order in the external voltage. Substituting the solution (3) into Eq. (1), the Fourier-transformed form of electric current is obtained,

$$\begin{aligned}I_\alpha(\omega)&=[\delta(\omega+\Omega)+\delta(\omega-\Omega)]\\ &\quad\times\sum_\beta G_{\alpha\beta}^{(\Omega)}(\omega)V_\beta+\delta(\omega)\sum_{\beta\gamma}G_{\alpha\beta\gamma}^{(0)}V_\beta V_\gamma\\ &\quad+[\delta(\omega+2\Omega)+\delta(\omega-2\Omega)]\sum_{\beta\gamma}G_{\alpha\beta\gamma}^{(2\Omega)}(\omega)V_\beta V_\gamma\\ &\quad+[\delta(\omega+\Omega)+\delta(\omega-\Omega)]\sum_{\beta\gamma\delta}G_{\alpha\beta\gamma\rho}^{(\Omega)}(\omega)V_\beta V_\gamma V_\rho\\ &\quad+[\delta(\omega+3\Omega)+\delta(\omega-3\Omega)]\sum_{\beta\gamma\delta}G_{\alpha\beta\gamma\rho}^{(3\Omega)}(\omega)V_\beta V_\gamma V_\rho\\ &\quad+\cdots.\end{aligned}\qquad(4)$$

The coefficients describing the total current flowing in and out of the conductor are a function of the unitary scattering matrix, which is influenced by the geometrical and intrinsic parameters of the system. From this relation it is found that in addition to the response at Ω, there are two components in the current for the lowest-order nonlinear response: one is a static term and another presents the oscillation at twice the driving frequency. The static component produces a dc electric current in the conductor, which corresponds to an optical rectification effect. The component oscillating at twice the driving frequency gives rise to second-harmonic generation. Third-harmonic generation occurs when an incident field at frequencies $\omega=\pm\Omega$ induces a response at frequencies $\omega=\pm3\Omega$. $G_{\alpha\beta}^{(\Omega)}(\omega)$ is a linear conductance that was first obtained by Büttiker and co-workers.[3–5] The quadratic conductance contains two contributions: a dc part $G_{\alpha\beta\gamma}^{(0)}=G_{\alpha\beta\gamma}^{\rm dc}$ and a part of the second-order harmonic generation $G_{\alpha\beta\gamma}^{(2\Omega)}(\omega)$

$=G^{\rm dc}_{\alpha\beta\gamma}+\omega(\mathrm{Re}\,G^{\rm ac}_{\alpha\beta\gamma}+i\,\mathrm{Im}\,G^{\rm ac}_{\alpha\beta\gamma})$, where

$$G^{\rm dc}_{\alpha\beta\gamma}=\frac{e^3}{2^2h}\int dE(-f'(E))\left\{\delta_{\beta\gamma}\partial_E A_{\beta\beta}(\alpha,E,E)+\int d\mathbf{r}\left[\left(u_\gamma\partial_U-\frac{du_\gamma}{dE}\right)A_{\beta\beta}(\alpha,E,E)+(\beta\leftrightarrow\gamma)\right]\right\},$$

$$\mathrm{Re}\,G^{\rm ac}_{\alpha\beta\gamma}=\frac{e^3}{2^5\pi}\int dE(-f'(E))\times\int d\mathbf{r}\left[\left(\frac{du_\gamma}{dE}\partial_U-\frac{d^2u_\gamma}{dE^2}\right)\times A_{\beta\beta}(\alpha,E,E)+(\beta\leftrightarrow\gamma)\right], \qquad (5)$$

$$\mathrm{Im}\,G^{\rm ac}_{\alpha\beta\gamma}=\frac{e^3}{2^3}\int dE(-f'(E))\int d\mathbf{r}(u_{\beta\gamma}\mathcal{F}_\alpha-\mathcal{F}_{\alpha\beta\gamma}).$$

The third-order conductance is given by $G^{(3\Omega)}_{\alpha\beta\gamma\rho}(\omega)=\frac{1}{3}G^{(\Omega)}_{\alpha\beta\gamma\rho}(\omega)=G^{\rm dc}_{\alpha\beta\gamma\rho}+\omega(\mathrm{Re}\,G^{\rm ac}_{\alpha\beta\gamma\rho}+i\,\mathrm{Im}\,G^{\rm ac}_{\alpha\beta\gamma\rho})$ with

$$G^{\rm dc}_{\alpha\beta\gamma\rho}=-\frac{e^4}{3!2^3h}\int dE(-f'(E))\Bigg(\delta_{\beta\gamma}\delta_{\beta\rho}\partial_E^2A_{\beta\beta}(\alpha,E,E)+\int d\mathbf{r}\Bigg\{\Bigg[u_{\gamma\rho}\partial_U+\delta_{\beta\gamma}u_\rho\partial_E\partial_U+u_\gamma u_\rho\partial_U^2-\delta_{\beta\gamma}\frac{d^2u_\rho}{dE^2}+\frac{d^2}{dE^2}(u_\gamma u_\rho)-\frac{du_{\gamma\rho}}{dE}+2\,\delta_{\beta\gamma}\frac{du_\rho}{dE}\partial_U-2\frac{d}{dE}(u_\gamma u_\rho)\partial_U\Bigg]A_{\beta\beta}(\alpha,E,E)\Bigg\}_c\Bigg),$$

$$\mathrm{Re}\,G^{\rm ac}_{\alpha\beta\gamma\rho}=\frac{e^4}{3!2^5\pi}\int dE(-f'(E))\int d\mathbf{r}\Bigg(\Bigg\{\frac{d^2u_{\gamma\rho}}{dE^2}-\frac{du_{\gamma\rho}}{dE}\partial_U+\delta_{\beta\gamma}\left(\frac{d^3u_\rho}{dE^3}-2\frac{d^2u_\rho}{dE^2}\partial_U-\frac{du_\rho}{dE}\partial_U\partial_E\right)-\left[u_\gamma\left(\frac{d^3u_\rho}{dE^3}-2\frac{d^2u_\rho}{dE^2}\partial_U+\frac{du_\rho}{dE}\partial_U^2\right)+(\gamma\leftrightarrow\rho)\right]-\left(4\frac{du_\gamma}{dE}\frac{d^2u_\rho}{dE^2}-2\frac{du_\gamma}{dE}\frac{du_\rho}{dE}+\frac{du_\gamma}{dE}u_\rho\right)+(\gamma\leftrightarrow\rho)\Bigg\}A_{\beta\beta}(\alpha,E,E)\Bigg)_c, \qquad (6)$$

$$\mathrm{Im}\,G^{\rm ac}_{\alpha\beta\gamma\rho}=\frac{e^4}{3!2^3}\int dE(-f'(E))\int d\mathbf{r}(u_{\beta\gamma\rho}\mathcal{F}_\alpha-\mathcal{F}_{\alpha\beta\gamma\rho}),$$

where the subscript c stands for the cyclic permutation of indices (except α). From Eqs. (5) and (6), it is found that in addition to the imaginary term $i\omega\,\mathrm{Im}\,G^{\rm ac}$ there exists the real term $\omega\,\mathrm{Re}\,G^{\rm ac}$, which changes the total current in the nonlinear response. Measurement of this effect would be an interesting subject for experimental studies. We also find that the internal interaction contributes to both the dc and the ac nonlinear conductances. Thus, the displacement current contributes to the total current. Employing the equations of characteristic tensors in Eq. (2) and those sum rules, all components of harmonic generation satisfy the current conservation and the gauge invariance.

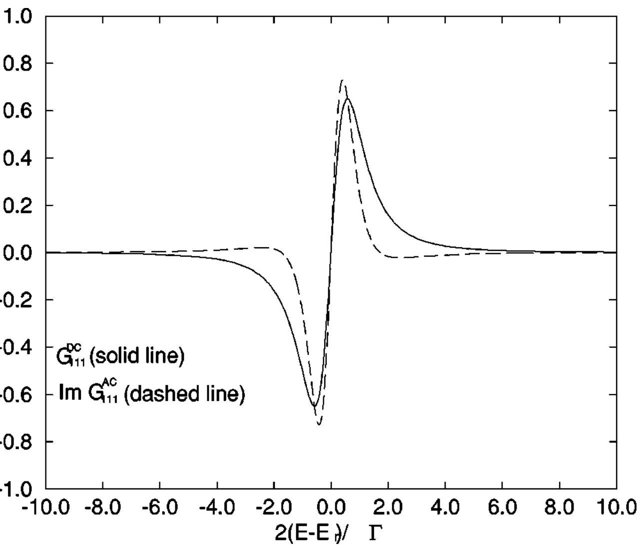

FIG. 1. The dc (solid line) and the ac (dashed line) quadratic conductances as the functions of $2(E-E_r)/\Gamma$ in units of $e^3\Gamma_1\Gamma_2(\Gamma_1-\Gamma_2)/\pi$. Set $\Gamma=1$ and $\hbar=1$.

As an example, we demonstrate the application of this formalism to a double-barrier tunneling diode. This system has been considered previously using the perturbation theory.[16] Consider a one-dimensional double-barrier tunneling system in which the two barriers are δ functions located at positions $x=-a$ and $x=a$. The scattering matrix close to the resonance is given by the Breit-Wigner formula $S_{\alpha\beta}(E)$

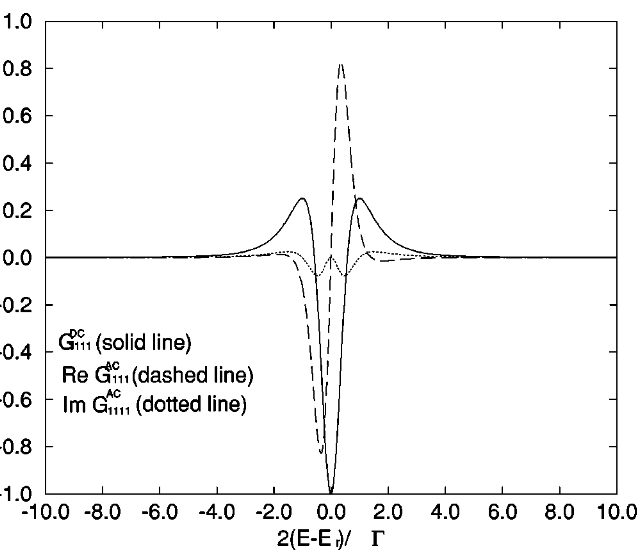

FIG. 2. $G^{\rm dc}_{1111}$ (solid line) in units of $e^4/3\times2^3\pi\hbar\Gamma^2$ and $G^{\rm ac}_{1111}$ as the functions of $2(E-E_r)/\Gamma$ in units of $e^4/2^3\pi\Gamma^3$. The dashed (dotted) line is the real (imaginary) part of $G^{\rm ac}_{1111}$.

$=[\delta_{\alpha\beta}-i\sqrt{\Gamma_\alpha\Gamma_\beta}/\triangle]e^{i(\phi_\alpha+\phi_\beta)}$ with $\triangle=E-E_r+i(\Gamma/2)$, where Γ_α ($\alpha=1,2$) are the partial widths of the resonances proportional to the tunneling probability through the barriers α and $\Gamma=\Sigma_\alpha\Gamma_\alpha$ is the total width of the resonance. ϕ_α is the phase acquired in the reflection or the transmission process. This allows simple expressions for the characteristic potential $u_1=\Gamma_1/\Gamma$ and $u_{11}=-2(\Gamma_1\Gamma_2/\Gamma^2)[(E-E_r)/|\triangle|^2]$. From Eq. (4), we obtain $G^{dc}_{111}=(e^3/2h)[(\Gamma_1-\Gamma_2)/\Gamma]\times(\Gamma_1\Gamma_2/|\triangle|^2)[(E-E_r)/|\triangle|^2]$, $\mathrm{Re}\,G^{ac}_{111}=0$ and $\mathrm{Im}\,G^{ac}_{111}(E)=-(e^3/2^3\pi)[(\Gamma_1-\Gamma_2)/\Gamma^2](\Gamma_1\Gamma_2/|\triangle|^2)\times[(|\triangle|^2-\Gamma^2)/|\triangle|^2][(E-E_r)/|\triangle|^2]$. The reason that $\mathrm{Re}\,G^{ac}_{111}$ vanishes in this example is the simple form of the Breit-Wigner approximation as it gives a space-independent and constant characteristic potential. $\mathrm{Re}\,G^{ac}_{111}$ would be nonzero in a space-dependent problem. In Fig. 1, G^{dc}_{111} (solid line) and $\mathrm{Im}\,G^{ac}_{111}$ (dashed line) are plotted as functions of $2(E-E_r)/\Gamma$. It is found that $G^{dc/ac}_{111}$ changes sign across the resonant point and hence can be negative.

For a geometrically symmetric double barrier, i.e., $\Gamma_1=\Gamma_2=\Gamma/2$, there is no quadratic conductance, i.e., $G^{dc/ac}_{111}=0$. The first nonzero nonlinear conductance is the third-order one. From Eq. (5), it is obtained $G^{dc}_{1111}(E)=(e^4/3!2^5h)[\Gamma^2/(|\triangle|^2)^2][(3|\triangle|^2-\Gamma^2)/|\triangle|^2]$, $\mathrm{Re}\,G^{ac}_{1111}(E)=-(e^4/2^{10}\pi)[\Gamma^2/(|\triangle|^2)^4](4|\triangle|^2-3\Gamma^2)(\triangle+\triangle^*)$, and $\mathrm{Im}\,G^{ac}_{1111}(E)=(e^4/2^{10}\pi)[\Gamma/(|\triangle|^2)^4][8(|\triangle|^2)^2-6|\triangle|^2\Gamma^2+\Gamma^4]$. G^{dc}_{1111} (solid line), $\mathrm{Re}\,G^{ac}_{1111}$ (dashed line), and $\mathrm{Im}\,G^{ac}_{1111}$ (dotted line) are plotted in Fig. 2 as a function of $2(E-E_r)/\Gamma$. Because $\mathrm{Re}\,G^{ac}_{1111}$ changes sign across the resonant point, it enhances or reduces the electric current. This effect would be small in the weakly nonlinear response.

In summary, harmonic generation in the ac nonlinear response has been formulated in the scattering matrix theory. The formalism takes into account the oscillating internal potential due to the charge redistribution. This allows us to obtain the response functions and the corresponding harmonic generation in a self-consistent way. Harmonic generation has been illustrated by obtaining the lowest-order and second-order nonlinear conductances. The internal interaction contributes to both the dc and the ac conductances in the regime of nonlinear response. The current conservation and gauge invariance are satisfied by all Fourier components of harmonic generation. It has been found that there exist frequency-dependent terms $\mathrm{Re}\,G^{ac}$ in the nonlinear conductances. As an example, the nonlinear conductances for a double-barrier nanostructure have been calculated. $\mathrm{Re}\,G^{ac}_{1111}$ has been calculated for a geometrically symmetric system. The quantum effect of $\omega\,\mathrm{Re}\,G^{ac}_{\mathrm{nonlinear}}$ might be experimentally observable in realistic structures.

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