

## Nonlinear transport theory for hybrid normal-superconducting devices

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We report a theory for analyzing nonlinear DC transport properties of mesoscopic or nanoscopic normal-superconducting (NS) systems. Special attention was paid such that our theory satisfies gauge invariance. At the nonlinear regime, our theory allows the investigation of a number of important problems: for NS hybrid systems we have derived the general nonlinear current-voltage characteristics in terms of the scattering Green's function, the second-order nonlinear conductance at the weakly nonlinear regime, and nonequilibrium charge pileup in the device that defines the electrochemical capacitance coefficients.

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### I. INTRODUCTION

The physics associated with quantum conduction in various low-dimensional hybrid superconducting systems has been a major focus of research at present.<sup>1-4</sup> Due to advances in controlled crystal growth and lithographic techniques, it is now possible to fabricate various submicron hybrid superconducting structures where accurate experimental measurements can be made.<sup>5</sup> An important theoretical task is to be able to predict, for general mesoscopic or nanoscopic hybrid systems, transport properties such as the nonlinear current-voltage characteristics and the nonequilibrium charge distribution inside the system as a function of the applied bias voltage. Our theoretical understanding of quantum transport in these very small normal superconducting (NS) hybrid systems has been achieved by scattering-matrix theory<sup>2,4</sup> and by non-equilibrium Green's function (NEGF) theory.<sup>6-10</sup>

To analyze nonlinear transport coefficients, i.e., coefficients that appear in front of nonlinear powers of bias voltage, in principle one must make sure gauge invariance of the theory. This means that theoretical results should not change when bias voltage applied at all the device leads is changed by the same amount. This is a necessary condition for any transport theory and has been recognized in the literature.<sup>11</sup> Consider a device that is connected to the outside world by several leads  $\alpha$  where bias voltage  $V_\alpha$  is applied. When  $V_\alpha \rightarrow V_\alpha + v$  where  $v$  is a constant, the calculated results (such as current) will not change if the electrostatic potential  $U$  inside the device is also changed by the same amount  $v$ . However  $U = U(\mathbf{r})$  which is the Hartree potential, can only be obtained by solving a self-consistent problem. In other words, to satisfy gauge invariance one necessarily requires to consider Coulomb interactions at least at the Hartree level. Furthermore, in general when external bias voltage is applied to a device, the flow of charge carriers through the device could polarize the system due to long-range Coulomb interactions. For a macroscopic metallic conductor, the polarization can be safely neglected since interaction is well screened. However for mesoscopic scale and nanoscale conductors the polarization could be very important. This also requires self-consistent analysis.

For *normal conductors*, Büttiker and his co-workers<sup>11,12</sup> developed an approach based on the scattering-matrix theory to deal with the second-order nonlinear conductance coefficients. This theory can also be extended to higher nonlinear order in DC situations.<sup>13</sup> On the other hand for a NS hybrid system, despite many theoretical investigations on its quantum-transport property,<sup>14</sup> the important issue of gauge invariance has not been clearly addressed so far. In light of this unsatisfactory situation, in this paper we report the development of a proper nonlinear-transport theory that satisfies gauge invariance for mesoscopic or nanoscopic NS hybrid device systems. Our theoretical formulation is based on nonequilibrium Green's function approach where the quantum-transport problem is solved in a self-consistent manner. We have formulated a gauge-invariant theory for the general I-V characteristics including the subgap behavior of the NS device in terms of Green's functions that are numerically calculable. In the weakly nonlinear regime we further derived the second order nonlinear coefficient by solving the characteristic potential. Since our theory can deal with charge polarization in the presence of transport,<sup>15,16</sup> we have also derived the linear and second-order nonlinear charge distributions that define the electrochemical capacitance of the system.

The rest of the paper is organized as follows. Section II presents the gauge-invariant nonlinear-transport theory for the NS system and Sec. III provides applications of our theory. Section IV is a short summary of this work.

### II. GAUGE-INVARIANT THEORY

In this section we formulate the gauge-invariant nonlinear-transport theory based on NEGF for NS hybrid device systems. To be specific, the NS system we consider is a quantum well connected to a normal metal lead and a superconducting lead. The current flowing through the normal lead is<sup>9</sup> ( $e = \hbar = 1$ ),

$$I = I_A + I_1, \quad (1)$$

with

$$I_A = 2 \int \frac{dE}{2\pi} \text{Tr}[\Gamma_L G_{12}^r \Gamma_L G_{12}^a][f_L(E + v_L - v_R) - f_L(E - v_L + v_R)], \quad (2)$$

$$I_1 = 2 \int \frac{dE}{2\pi} \rho_R(E) \text{Tr} \left[ \Gamma_L G_{11}^r \Gamma_R G_{11}^a + \Gamma_L G_{12}^r \Gamma_R G_{12}^a - \frac{\Delta}{|E|} (\Gamma_L G_{11}^r \Gamma_R G_{12}^a + \Gamma_L G_{12}^r \Gamma_R G_{11}^a) \right] \times [f_L(E + v_L - v_R) - f_R(E)], \quad (3)$$

where  $G_{11}$  and  $G_{12}$  are the matrix elements of the  $2 \times 2$  Nambu representation. Here

$$G_{11}^r(E) = [E - H_d + U - v_R - \Sigma_{11}^r - \Sigma_{12}^r A^r \Sigma_{21}^r]^{-1} \quad (4)$$

and

$$A^r = [E + H_d^* + v_R - U - \Sigma_{22}^r]^{-1}. \quad (5)$$

Once the electron and hole Green's functions  $G_{11}^r$  and  $A^r$  were obtained,  $G_{12}^r$  is calculated by

$$G_{12}^r = G_{11}^r \Sigma_{12}^r A^r, \quad (6)$$

where  $\Sigma^r$  is the self-energy.

We emphasize that the crucial step in developing the gauge-invariant nonlinear DC theory is to include the *internal* potential landscape  $U(\mathbf{r})$  into the Green's functions self-consistently.<sup>17</sup> In this work we deal with it at the Hartree level, hence  $U(\mathbf{r})$  is determined by the self-consistent Poisson equation

$$\nabla^2 U(x) = -4\pi i (G_{11}^<(E, U))_{xx} \quad (7)$$

where  $G_{11}^<$  is the electron lesser Green's function in real space and  $x$  labels the three-dimensional position. From Ref. 9 we have

$$G_{11}^< = i \int \frac{dE}{2\pi} [G_{11}^r \Gamma_L G_{11}^a f_L(E + v_L - v_R) + G_{12}^r \Gamma_L G_{21}^a f_L(E - v_L + v_R)] + \int \frac{dE}{2\pi} \rho_R(E) f_R(E) \left[ G_{11}^r \Gamma_R G_{11}^a + G_{12}^r \Gamma_R G_{21}^a - \frac{\Delta}{|E|} (G_{11}^r \Gamma_R G_{21}^a + G_{12}^r \Gamma_R G_{11}^a) \right]. \quad (8)$$

Equations (2), (3), and (7) completely determine the nonlinear I-V characteristics of the NS hybrid system: they form the basic equations of the gauge-invariant nonlinear theory. The self-consistent nature of the problem is clear: one must solve the quantum-scattering problem (the Green's functions) in conjunction with the Poisson equation. It is easy to prove that the current expression Eqs. (2) and (3) are gauge invariant. Equations (2), (3), and (7) also form the basis for numerical analysis of I-V curves for the NS system. For instance one can compute the various Green's functions  $G$

and the coupling matrix  $\Gamma$  using tight-binding models,<sup>8</sup> and the Poisson equation can be efficiently solved in real space by powerful numerical techniques.<sup>16</sup>

In the simplest approximation, the gauge-invariant condition can be satisfied by putting a gate voltage  $V_g$  as was done in Ref. 9 so that one treats the system as a three-probe conductor with external voltages  $V_L$ ,  $V_R$ , and  $V_g$  applied at the probes. In general, the internal potential is a nonlinear function of  $V_\alpha$  (see Sec. IV for details), but as a first approximation one expands it in terms of  $V_\alpha$  in the small voltage limit,

$$U = u_L V_L + u_R V_R + u_g V_g \quad (9)$$

where  $u_\alpha(\mathbf{r})$  is the characteristic potential that satisfies the sum rule  $\sum_\alpha u_\alpha = 1$ . If one makes<sup>9</sup> a further approximation by assuming  $u_L = u_R = 0$ , the sum rule gives  $u_g = 1$  and  $U = V_g$ , i.e.,  $U$  is just a constant-potential shift under these approximations.

In distinct contrast to the constant  $U$  model, the theory presented in this section is a microscopic gauge-invariant theory. Furthermore, in order to discuss charge polarization and electrochemical capacitance in the presence of transport, one has to include the self-consistent Hartree field rather than just include a constant gate voltage: one can easily confirm that the constant  $U$  model corresponds to the local-charge-neutrality approximation, it will therefore not give rise to any charge polarization.

### III. APPLICATIONS

In this section we present detailed analysis of situations where analytical expressions can be obtained in closed form. These are the second-order weakly nonlinear conductance and the nonequilibrium charge distribution.

#### A. Weakly nonlinear regime

For weak nonlinearity we can expand all quantities in terms of the external bias voltage<sup>11</sup> and obtain results order-by-order. Such an expansion makes sense when bias is finite but small. This approach was adapted in SMT (Ref. 11) and response theory<sup>13</sup> for analyzing normal mesoscopic conductors. For the NS system we will derive formula for the local density of states (LDOS) and the second-order weakly nonlinear DC conductance. These are the interesting quantities for weakly nonlinear regime.

In both SMT (Ref. 11) and response theory,<sup>13</sup> LDOS plays a very important role. From our NEGF theory LDOS can be easily derived from the right-hand side of Eq. (7), which is the charge density, with the help of Eq. (8). Here we shall present the explicit expression at the lowest-order<sup>13</sup> expansion in external bias. Hence we seek the solution of  $U(\mathbf{r})$  in the following form,

$$U = \sum_\alpha u_\alpha v_\alpha + \frac{1}{2} \sum_{\alpha\beta} u_{\alpha\beta} v_\alpha v_\beta + \dots \quad (10)$$

where  $u_\alpha(\mathbf{r})$  and  $u_{\alpha\beta\dots}(\mathbf{r})$  are the characteristic potentials.<sup>11,13</sup> It can be shown that the characteristic potential satisfies many sum rules,<sup>11,13</sup>  $\sum_\alpha u_\alpha = 1$  and  $\sum_{\gamma \in \beta} u_{\alpha\{\beta\}_\gamma}$

$=0$ , where the subscript  $\{\beta\}_l$  is a short notation of  $l$  indices  $\gamma, \delta, \eta, \dots$ . Expanding  $G_{11}^<$  of Eq. (7) in power series of  $v_\alpha$ ,<sup>18</sup> we can derive equations for all the characteristic potentials. In particular the expansions are facilitated by iterating the following Dyson equation to the appropriate order

$$A^r = A_0^r - A^r(v_R - U)A_0^r$$

and

$$G_{11}^r = G_{11,0}^r - G_{11}^r(U - v_R)G_{11,0}^r + G_{11}^r \Sigma_{12}^r (A^r - A_0^r) \Sigma_{21}^r G_{11,0}^r, \quad (11)$$

where  $A_0^r$  and  $G_{11,0}^r$  are equilibrium hole and electron Green's functions. The expansion of  $G_{12}^r$  can be made similarly. At the lowest order, we thus obtain the local charge density in the presence of transport,<sup>19</sup>

$$\rho(x) = i(G_{11}^< - G_{11,0}^<)_{xx} = \rho_{inj} + \rho_{ind}, \quad (12)$$

where

$$\rho_{inj} = (dn_e/dE - dn_h/dE)(v_L - v_R) - (1/2)(d^2n_e/dE^2 + d^2n_h/dE^2)(v_L - v_R)^2 \quad (13)$$

is the injected charge from the normal lead.  $dn_e/dE$  is the injectivity of electron, i.e., the DOS for an electron coming from left lead and exiting the system as an electron,

$$dn_e(x)/dE = \int (dE/2\pi)(-\partial_E f_L)(G_{11,0}^r \Gamma_L G_{11,0}^a)_{xx}. \quad (14)$$

In addition  $dn_h/dE$  is the injectivity of a hole, i.e., the DOS for a hole coming from left lead and exiting the system as an electron,

$$dn_h(x)/dE = \int (dE/2\pi)(-\partial_E f_L)(G_{12,0}^r \Gamma_L G_{12,0}^a)_{xx}. \quad (15)$$

Finally  $d^2n/dE^2$  is the derivative of  $dn/dE$  with respect to energy. Note that Eqs. (14) and (15) are the same as that defined in the scattering approach of Gramespacher and Büttiker.<sup>20</sup>

In Eq. (12) the induced charge due to long-range Coulomb interactions is derived to be given by

$$\begin{aligned} \rho_{ind}(x) &= - \int (dE/\pi) f_L \\ &\quad \times \text{Im}[G_{11,0}^r(u_L - \Sigma_{12}^r A_0^r u_L A_0^r \Sigma_{21}^r) G_{11,0}^r]_{xx} \\ &\quad (v_L - v_R) \\ &\equiv - \sum_{x'} \Pi_{xx'} u_L(x') (v_L - v_R), \end{aligned} \quad (16)$$

where  $\Pi$  is the generalized Lindhard function that reduces to the Lindhard function of normal conductor<sup>21,11,13</sup> in the limit  $\Delta \rightarrow 0$ . For example, using the wide bandwidth limit<sup>22</sup> expressions for the Green's function  $G_{11}^r$  at small Fermi energy we can calculate the Lindhard function  $\Pi$  exactly at zero temperature from its definition above:

$$\Pi = \frac{1}{\pi \Gamma_R} \left[ \frac{\pi}{2} - \arctan \frac{2E^2 - (\Gamma_R^2 - \Gamma_L^2)/2}{\Gamma_L \Gamma_R} \right], \quad (17)$$

where we have set the quantum-well level  $E_o = 0$ . Hence the Lindhard function is a smooth function increasing with energy monotonically.

With these quantities the Poisson equation becomes,

$$-\nabla^2 u_L(x) + 4\pi \sum_{x'} \Pi_{xx'} u_L(x') = 4\pi \left( \frac{dn_e(x)}{dE} - \frac{dn_h(x)}{dE} \right) \quad (18)$$

$$\begin{aligned} &-\nabla^2 u_{LL}(x) + 4\pi \sum_{x'} \Pi_{xx'} u_{LL}(x') \\ &= 4\pi \left( \frac{d\tilde{n}_e(x)}{dE} - \frac{d\tilde{n}_h(x)}{dE} \right), \end{aligned} \quad (19)$$

where  $d\tilde{n}_e/dE$  and  $d\tilde{n}_h/dE$  are the second-order injectivities.<sup>13,23</sup> These partial-differential equations can at least be solved numerically. However to avoid numerics one may apply the quasineutrality approximation<sup>12</sup> by neglecting the spatial derivative in Eq. (18). This way the characteristic potential is obtained as<sup>24,25</sup>

$$u_L = \left( \frac{dn_e}{dE} - \frac{dn_h}{dE} \right) / \Pi. \quad (20)$$

In terms of the characteristic potential we now derive the second-order nonlinear conductance due to Andreev reflection. In the weakly nonlinear regime, only the Andreev current  $I_A$  is relevant that can be expanded in terms of external bias voltage difference  $v \equiv v_L - v_R$ ,

$$I_A = G_{11}v + G_{111}v^2 + \dots$$

From this definition of conductance coefficients  $G_{11}$  and  $G_{111}$ , we expand Eq. (2) in terms of  $v$  to obtain,

$$G_{11} = 4 \int (dE/2\pi)(-\partial_E f_L) T_A$$

and

$$G_{111} = -4 \int \frac{dE}{2\pi} (-\partial_E f_L) \text{Tr} \left[ \frac{dG_A}{dU} u_L \right] \quad (21)$$

where  $G_A \equiv \Gamma_L G_{12}^r \Gamma_L G_{12}^a$  and  $dG_A/dU$  is easily calculable using Eq. (11) and the relation in Ref. 19. To compare with the second-order conductance of normal conductor  $G_{111}^N$ , we note that  $G_{111}^N$  has two contributions.<sup>12,17</sup> One of them comes from Coulomb interaction,

$$\begin{aligned} G_{111}^N &= \int (dE/2\pi) \text{Tr} [G_0^a (\Gamma_L G_0^r u_L + u_L G_0^a \Gamma_L - 1/2 \Gamma_L G_0^r \\ &\quad - 1/2 G_0^a \Gamma_L) G_0^r \Gamma_R] \partial_E f. \end{aligned} \quad (22)$$

However, for NS system, if the Coulomb interaction is not important (when  $u_L = 0$ ), we would have  $G_{111} = 0$ . For example, for an ideal ballistic wire, or for a symmetric quan-

tum well at a resonant tunneling point, every incident charge is perfectly Andreev reflected. Therefore, for these examples no charge accumulation is possible. From Eqs. (14) and (15), we can easily verify that  $dn_e/dE = dn_h/dE$  near a resonant point for a symmetric system and hence a vanishing  $G_{111}$  since  $u_L = 0$  from Eq. (20). In contrast, when  $u_L = 0$ ,  $G_{111}^N$  is nonzero from Eq. (22).

### B. Electrochemical capacitance

Using the NEGF theory one can also investigate the non-equilibrium charge distribution inside the NS system. For this purpose we divide the system into two regions: in region I the charge is positive and in region II it is negative. The total charge in region I can be calculated using Eq. (12):  $Q_I = \int_I \rho(x) dx$ . Expanding  $Q_I$  in powers of  $v$  in the following form<sup>13,23</sup>

$$Q_I = C_{11}v + \frac{1}{2}C_{111}v^2 + \dots \equiv C(v)$$

this defines the electrochemical capacitance coefficients  $C_{11}$ ,  $C_{111}$ , and the general voltage dependent electrochemical capacitance<sup>26</sup>  $C(v)$ . It is not difficult to confirm that the first two coefficients are

$$C_{11} = \int_I dx \left( \frac{dn_e}{dE} - \frac{dn_h}{dE} \right) - \int_I dx dx' \Pi(x, x') u_L(x') \quad (23)$$

$$C_{111} = \int_I dx \left( \frac{d\tilde{n}_e}{dE} - \frac{d\tilde{n}_h}{dE} \right) - \int_I dx dx' \Pi(x, x') u_{LL}(x'). \quad (24)$$

To get some physical insight into these coefficients we consider the discrete potential model.<sup>15</sup> In addition, we parametrize the characteristic potentials by the geometric capacitance  $C_0$ , in terms of which the Poisson equation becomes

$$C_0(u_I - u_{II}) = (D_I^e - D_I^h)v - \Pi_I u_I = C_{11}v, \quad (25)$$

$$-C_0(u_I - u_{II}) = (D_{II}^e - D_{II}^h)v - \Pi_{II} u_{II}, \quad (26)$$

where we have set  $D^e = \int_I dx (dn_e/dE)$ ,  $D^h = \int_I dx (dn_h/dE)$ ,  $\Pi_I = \int_I dx \Pi(x, x)$  and small bias limit is assumed. We solve the characteristic potentials  $u_L$  and  $u_{LL}$  through these two equations in terms of  $C_0$ . This leads to the following expression for the electrochemical capacitance coefficient  $C_{11}$  for a NS system:

$$C_{11} = \frac{(D_I^e - D_I^h)/\Pi_I - (D_{II}^e - D_{II}^h)/\Pi_{II}}{C_0^{-1} + \Pi_I^{-1} + \Pi_{II}^{-1}}. \quad (27)$$

In particular, in the limit of gap  $\Delta \rightarrow 0$ , from Eqs. (14), (15), and (16) we obtain  $D^h = 0$ ,  $\Pi = dn/dE$ , and  $D^e = dn_L/dE$  where  $dn_L/dE$  is the injectivity of left lead. In this situation Eq. (27) reduces to the expression of the electrochemical capacitance for a normal conductor.<sup>23</sup> Let us consider a symmetric-tunneling NS system. At the resonant point, the electron will be reflected as the hole due to the Andreev reflection. As a result, the capacitance  $C_{11}$  vanishes since  $D^e = D^h$  and there is no charge accumulation.

### IV. SUMMARY

In this work we have developed a gauge-invariant NEGF theory for hybrid NS systems. This theory explicitly takes into account the long-range Coulomb interaction in the normal region. Because of gauge invariance, our theory is applicable for nonlinear regime for which we have derived an explicit expression for nonlinear current-voltage characteristics for NS devices. This result can be further simplified in the weakly nonlinear regime, for which we have analyzed the second-order nonlinear conductance and the generalized Lindhard function. It is interesting to see that for NS systems the concept of injectivity is naturally extended to include the injectivity of holes: these quantities automatically appear in our formalism. Our theory included charge-polarization effect hence can be applied to analyze the electrochemical capacitance coefficients at the linear and nonlinear orders in bias. In particular we have derived an analytic expression of the linear electrochemical capacitance of NS system within the discrete potential model.

While this paper concentrated on the development of a theoretical formalism in terms of the Green's functions, it is obvious that numerical computations can be carried out applying the analytical expressions derived here. This way one can avoid the various approximations used here in order to obtain closed form results. Of particular interests are the investigation of nonlinear I-V curves without the wide-bandwidth approximation; the calculation of nonlinear conductance coefficients without the quasineutrality approximation; and the study of nonequilibrium charge distribution without the discrete potential approximation. These, however, will be the subject of a future report.

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