PHYSICAL REVIEW B VOLUME 38, NUMBER 15 15 NOVEMBER 1988-II

Nonequilibrium Green's functions and nonlinear quantum transport coefficients

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(Received 23 May 1988)

Nonlinear quantum transport coefficients are discussed in terms of nonequilibrium Green's functions, and are finally expressed in terms of retarded Green's functions alone for the case of impurity scattering. These results are well adapted to describe nonlinear steady state dc- and ac- as well as transient-current response (linear and bilinear) to time-dependent and spatially inhomogeneous electric fields, and can also accommodate an ambient magnetic field.

I. INTRODUCTION AND FORMULATION

This work is concerned with the examination of an alternative procedure for the construction of nonequilibrium Green's functions appropriate to the analysis of nonlinear quantum transport theory. Our considerations here are focused on the case where the only electron scatterings are due to impurity scattering centers at fixed sites. This constitutes a genuinely one-particle dynamical situation as there is no coupling of the second quantized electron field operator $\psi(\mathbf{x},t)$ to any other dynamical field, and its time development is governed by ($\hbar \rightarrow 1$, $c \rightarrow 1$)

$$\left\{ i\frac{\partial}{\partial t}+\frac{1}{2m}[\nabla - ie\mathbf{a}(\mathbf{x})-ie\,\mathbf{A}_e(\mathbf{x},t)]^2 - [U(\mathbf{x})+V_e(\mathbf{x},t)]\right\}\psi(\mathbf{x},t)=0 \;, \qquad (1)$$

where $\mathbf{a}(\mathbf{x})$ is the vector potential of a time-independent ambient magnetic field, $U(\mathbf{x})=\sum_i V(\mathbf{x}-\mathbf{r}_i)$ is the potential of impurity scatterers centered at $\mathbf{r}_i$, and $\mathbf{A}_e(\mathbf{x},t)$ and $V_e(\mathbf{x},t)$ are electromagnetic vector and scalar potentials for fields which drive the system out of equilibrium. We note that the time development of $\psi(\mathbf{x},t)$ associated with single-particle dynamics is described by

$$\psi(\mathbf{x},t)=i\int d^3x' G^r(\mathbf{x},t;\mathbf{x}',t')\psi(\mathbf{x}',t') \;, \qquad (2)$$

where the retarded Green's function $G^r(\mathbf{x},t;\mathbf{x}',t')$ satisfies the equation

$$\left\{ i\frac{\partial}{\partial t}+\frac{1}{2m}[\nabla - ie\mathbf{a}(\mathbf{x})-ie\,\mathbf{A}_e(\mathbf{x},t)]^2-[U(\mathbf{x})+V_e(\mathbf{x},t)]\right\} \times G^r(\mathbf{x},t;\mathbf{x}',t')=\delta^3(\mathbf{x}-\mathbf{x}')\delta(t-t') \;, \qquad (3)$$

subject to the initial retardation conditions $G^r(\mathbf{x},t;\mathbf{x}',t'>t)=0$ and $G^r(\mathbf{x},t^+;\mathbf{x}',t)=-i\delta^3(\mathbf{x}-\mathbf{x}')$. It is readily verified that for $t>t'$, $\psi(\mathbf{x},t)$ given by Eq. (2) satisfies Eq. (1), and that for $t=t'+\varepsilon$, Eq. (2) correctly reproduces the initial value; moreover, in its role as time-development operator $S(t,t')$ we have

$$G^r(\mathbf{x},t;\mathbf{x}',t')=-i\langle \mathbf{x}\,|\,S(t,t')\,|\,\mathbf{x}'\rangle \qquad (4)$$

for single-particle dynamics. Moreover, the retarded Green's-function description of time development given by Eq. (2) correctly propagates the equal-time canonical commutation relations forward in time as

$$[\psi(\mathbf{x},t),\psi^\dagger(\mathbf{x}',t)]_\mp=[\psi(\mathbf{x},0),\psi^\dagger(\mathbf{x}',0)]_\mp =\delta^3(\mathbf{x}-\mathbf{x}') \;, \qquad (5)$$

($\mp$ denotes commutation and anticommutation relations for bosons and fermions). In transport, the focus is on the current density operator

$$\mathbf{j}(\mathbf{x},t)=\frac{e}{2m}\{\, i[\nabla\psi^\dagger(\mathbf{x},t)]\psi(\mathbf{x},t)-\psi^\dagger(\mathbf{x},t)i\nabla\psi(\mathbf{x},t) -2e[\,\mathbf{A}_e(\mathbf{x},t)+\mathbf{a}(\mathbf{x})]\psi^\dagger(\mathbf{x},t)\psi(\mathbf{x},t)\} \;, \qquad (6)$$

which we consider to be averaged in an initial equilibrium ensemble $\exp[-\beta(H_0-\zeta N)]$, where the initial Hamiltonian H_0 contains $\mathbf{a}(\mathbf{x})$ and $U(\mathbf{x})$ [before $\mathbf{A}_e(\mathbf{x},t)$ and $V_e(\mathbf{x},t)$ are turned on], N is the number operator, and β and ζ are the inverse thermal energy $1/k_BT$ and chemical potential of the initial equilibrium state, respectively. Such initial equilibrium ensemble averaging of an operator X is given by

$$\langle X\rangle=\mathrm{Tr}(e^{-\beta(H_0-\zeta N)}X)/\mathrm{Tr}e^{-\beta(H_0-\zeta N)} \qquad (7)$$

and the associated physical nonequilibrium Green's function is given by (Wick time ordering is indicated by the symbol T, which we understand to include $\pm$ accompanying the "lesser" time order for bosons and fermions)

$$g(\mathbf{x},t;\mathbf{x}',t')=-i\langle\, T[\psi(\mathbf{x},t)\psi^\dagger(\mathbf{x}',t')]\rangle \;, \qquad (8)$$

with

$$g_<(\mathbf{x},t;\mathbf{x}',t')=i\langle\,\psi^\dagger(\mathbf{x}',t')\psi(\mathbf{x},t)\rangle \;,$$

etc. Of course the time development of $\psi(\mathbf{x},t)$ is governed by the full Hamiltonian including $\mathbf{A}_e(\mathbf{x},t)$ and $V_e(\mathbf{x},t)$, not H_0 associated with the initial equilibrium averaging. The average current $\langle\,\mathbf{j}(\mathbf{x},t)\rangle$ which evolves by the action of $\mathbf{A}_e(\mathbf{x},t)$ and $V_e(\mathbf{x},t)$ driving the system out of the initial equilibrium state is given by

$$\langle\,\mathbf{j}(\mathbf{x},t)\rangle=-\frac{e}{2m}\lim_{\mathbf{x}'\rightarrow\mathbf{x}}\{\nabla-\nabla'-2ie[\,\mathbf{A}_e(\mathbf{x},t)+\mathbf{a}(\mathbf{x})]\} \times g_<(\mathbf{x},t;\mathbf{x}',t) \;. \qquad (9)$$

We note that we may rewrite Eq. (8) using Eq. (2) as

$$g_<(\mathbf{x},t;\mathbf{x}',t')=\int d^3x_1\int d^3x_2 G^r(\mathbf{x},t;\mathbf{x}_1 0)g_<^{(0)}(\mathbf{x}_1,0;\mathbf{x}_2,0)G^{r*}(\mathbf{x}',t';\mathbf{x}_2,0)\ , \tag{10}$$

where the retarded Green's function G^r has been employed to refer the time arguments of ψ, $\psi^\dagger$, and g back to the initial time (0) when the system is in equilibrium, and the initial equilibrium Green's function is denoted by $g^{(0)}$. Equation (10) has the interesting property that the highly complex problem of nonequilibrium statistical mechanics (the determination of $g_<$), which involves statistical and dynamical considerations *jointly* in nonequilibrium processes, is divided by Eq. (10) into two distinct and separated parts such that only one part involves statistical averaging in the context of the equilibrium Green's function $g^{(0)}$, while the other part is devoid of statistical averaging, and involves only single-particle dynamics in external fields in the context of the retarded Green's function G^r, G^{r*}. This is to say that Eq. (10) serves to break down nonequilibrium statistical mechanics into the following parts—to all orders in the external fields—(1) a problem of equilibrium statistical mechanics (the determination of $g^{(0)}$) and (2) a problem of nonlinear single-particle dynamics in external fields, devoid of statistical averaging (the determination of G^r). (Scatterings are present in both $g^{(0)}$ and G^r.) The formulation of Eq. (10) is particularly useful if the fields of $\mathbf{A}_e(\mathbf{x},t)$ and $V_e(\mathbf{x},t)$ are abruptly turned on at initial time 0, and we wish to trace the time development of transient currents under conditions where there is no transfer of energy to a heat bath. It should be noted that while $g(\mathbf{x},t;\mathbf{x}',t')$ and $G^r(\mathbf{x},t;\mathbf{x}',t')$ obey the same equation of motion [both obey Eq. (3)], their difference in initial conditions is crucial in determining the difficulty of solution—G^r is subject to the initial condition of retardation, whereas g is subject to initial equilibrium state averaging. In the special case when $\mathbf{A}_e(\mathbf{x},t)$ and $V_e(\mathbf{x},t)$ vanish identically, g is an equilibrium thermal Green's function[1] at all times, with initial Hamiltonian H_0 identical to the Hamiltonian generator of time translations, and this yields the characteristic periodicity-antiperiodicity conditions for time-difference variables with imaginary period $i\beta$ leading to the appearance of Fermi-Bose distribution functions in the spectral structure of $g^{(0)}$. With time-variable fields turned on, H_0 is no longer the generator of time translations, and greater complexities of g in regard to its initial equilibrium state averaging have been described in the literature.[2–10] An exact relation of the physical nonequilibrium Green's function g to a corresponding "generating" Green's function employed by Martin and Schwinger[1–3] has been derived by Kadanoff and Baym,[3] and it has been employed in the development of transport equations which embody the difficulties of determining g (which difficulties also permeate the corresponding formulations of perturbation theory for nonequilibrium Green's functions).[3–10] The "generating" Green's function replaces the canonical weight factor $\exp(-\beta H_0)$ by the time-ordered exponential which actually generates time displacements through $i\beta$ in time-dependent external fields, and thus has periodicity properties in each time index separately. This generating Green's function is given in the full Heisenberg picture by

$$G(\mathbf{x},t;\mathbf{x}',t';t_0)=\frac{-i\,\mathrm{Tr}\{S(t_0-i\beta,t_0)T[\psi(\mathbf{x},t)\psi^\dagger(\mathbf{x}',t')]\}}{\mathrm{Tr}[S(t_0-i\beta,t_0)]}\ , \tag{11}$$

which is equivalent to the more frequently used interaction-picture form (corresponding to the separation of the time-dependent external field Hamiltonian $H_e=H-H_0$ associated with $\mathbf{A}_e$ and V_e)

$$G(\mathbf{x},t;\mathbf{x}',t';t_0)=\frac{-i\,\mathrm{Tr}\{T[S(t_0-i\beta,t_0)\psi_I(\mathbf{x},t)\psi_I^\dagger(\mathbf{x}',t')]\}}{\mathrm{Tr}[S(t_0-i\beta,t_0)]}\ , \tag{12}$$

where the subscript I indicates interaction-picture operators and subscript S will indicate Schrödinger-picture operators (and all operators with no such subscripts are of the full Heisenberg picture). Suppressing the explicit appearance of the chemical potential as energy reference level, the imaginary time-displacement operator $S(t_0-i\beta,t_0)$ has the equivalent forms

$$\begin{aligned}S(t_0-i\beta,t_0)&=\left[\exp\left[-i\int_{t_0}^{t_0-i\beta}dt'H_S(t')\right]\right]_+\\&=\left[\exp\left[-i\int_{t_0}^{t_0-i\beta}dt'H(t')\right]\right]_-\\&=\exp(-\beta H_{0I})\left[\exp\left[-i\int_{t_0}^{t_0-i\beta}dt'H_{eI}(t')\right]\right]_+\ ,\end{aligned} \tag{13}$$

where the subscripts $(+,-)$ mean time ordering in the (positive, negative) sense. The imaginary time-periodicity properties of the generating Green's function have been demonstrated by Ashby[2] and Kadanoff and Baym,[3] in the form

$$\begin{aligned}G_<(\mathbf{x},t_0;\mathbf{x}',t';t_0)&=\pm G_>(\mathbf{x},t_0-i\beta;\mathbf{x}',t';t_0)\ ,\\G_>(\mathbf{x},t;\mathbf{x}',t_0;t_0)&=\pm G_<(\mathbf{x},t;\mathbf{x}',t_0-i\beta;t_0)\ .\end{aligned} \tag{14}$$

It is well known[1–3] that $G(\mathbf{x},t;\mathbf{x}',t';t_0)$ has the useful property[1-3] that its variational derivatives with respect to external potential generate higher-order nonequilibrium generating Green's functions of the same type, and its relation to the physical nonequilibrium Green's function $g(\mathbf{x},t;\mathbf{x}',t')$—which lacks such a convenient variational differential property—is given by

$$\lim_{t_0\to-\infty} G(\mathbf{x},t;\mathbf{x}',t';t_0)=g(\mathbf{x},t;\mathbf{x}',t') \ . \tag{15}$$

Notwithstanding the attractive features of the generating Green's function G, our focus is upon the physical nonequilibrium Green's function g directly. The present decomposition of the problem of nonequilibrium statistical mechanics into one of equilibrium statistical mechanics and another of nonlinear single-particle dynamics devoid of statistical averaging for the case of impurity scattering (single-particle dynamics)—as represented by Eq. (10)—offers an alternative point of view which may hold some advantage in dealing with the severe complications cited above due to the fact that there is no corresponding problem of dealing with the initial condition on G^r, as it is simply one of retardation. With this in view we rewrite Eq. (3) as an integral equation

$$\begin{aligned} G^r(\mathbf{x},t;\mathbf{x}',t')&=G_0^r(\mathbf{x},t;\mathbf{x}',t') \\ &\quad-\int d^3x''\int dt''G_0^r(\mathbf{x},t;\mathbf{x}'',t'') \\ &\quad\times X_{\text{op}}(\mathbf{x}'',t'')G^r(\mathbf{x}'',t'';\mathbf{x}',t') \ , \end{aligned} \tag{16}$$

where G_0^r is the retarded Green's function in the absence of $\mathbf{A}_e$ and V_e. Taking the gauge in which $V_e\to 0$, $X_{\text{op}}(\mathbf{x},t)$ is given by $X_{\text{op}}(\mathbf{x},t)=X_{\text{op}}^{(1)}(\mathbf{x},t)+X^{(2)}(\mathbf{x},t)$ where

$$\begin{aligned} X_{\text{op}}^{(1)}(\mathbf{x},t)&=\frac{ie}{2m}[\nabla\cdot\mathbf{A}_e(\mathbf{x},t)]+\frac{ie}{m}\mathbf{A}_e(\mathbf{x},t)\cdot\nabla \ , \\ X^{(2)}(\mathbf{x},t)&=-\frac{e^2}{2m}A_e^2(\mathbf{x},t) \ . \end{aligned} \tag{17}$$

[Note that $\nabla\cdot\mathbf{A}_e(\mathbf{x},\omega)\sim\nabla\cdot\mathbf{E}(\mathbf{x},\omega)\sim\rho_e(\mathbf{x},\omega)\to 0$ for impressed fields due to far-off sources ρ_e in frequency representation.]

II. LINEAR AND BILINEAR CONDUCTIVITY COEFFICIENTS IN TERMS OF RETARDED GREEN'S FUNCTIONS

Iteration of Eq. (16) may be used in conjunction with Eqs. (9) and (10) to generate nonlinear transport coefficients which are variants of Kubo-type response formulas[11] generalized to nonlinear response[12-14] and expressed in terms of retarded Green's functions [note that $X_{\text{op}}^{(1,2)}(\mathbf{x},t)$ is simply related to the electric field in frequency-ω representation since $\mathbf{E}(\omega)=i\omega\,\mathbf{A}_e(\omega)$]. For example, to second order in the field [$1=(\mathbf{x},t)$, $\int d^41=\int d^3x\int dt$],

$$\begin{aligned} G^r(1,1')=&\ G_0^r(1,1')-\int d^42\,G_0^r(1,2)X_{\text{op}}^{(1)}(2)G_0^r(2,1')-\int d^42\,G_0^r(1,2)X^{(2)}(2)G_0^r(2,1') \\ &+\int d^42\int d^43\,G_0^r(1,2)X_{\text{op}}^{(1)}(2)G_0^r(2,3)X_{\text{op}}^{(1)}(3)G_0^r(3,1') \ . \end{aligned} \tag{18}$$

In employing Eq. (18) to construct the nonequilibrium Green's function of Eq. (10), $g_<(1,1')$, we use the identity representing the propagation of the *equilibrium* Green's function forward in time as

$$g_<^{(0)}(\mathbf{x},t;\mathbf{x}',t')=\int d^3x_1\int d^3x_2G_0^r(\mathbf{x},t;\mathbf{x}_1,0)g_<^{(0)}(\mathbf{x}_1,0;\mathbf{x}_2,0)G_0^{r*}(\mathbf{x}',t';\mathbf{x}_2,0) \tag{19}$$

and in passing we note that the equilibrium Green's function is given by[1] [$f_0(\omega)$ is the Fermi distribution function]

$$g_<^{(0)}(\mathbf{x},t;\mathbf{x}',t')=-\int\frac{d\omega}{2\pi}e^{-i\omega(t-t')}f_0(\omega)a_0(\mathbf{x},\mathbf{x}';\omega) \ , \tag{20a}$$

where the spectral weight function $a_0(\mathbf{x},\mathbf{x}';\omega)$ is given by

$$a_0(\mathbf{x},\mathbf{x}';\omega)=G_0^r(\mathbf{x},\mathbf{x}';\omega+i\epsilon)-G_0^r(\mathbf{x},\mathbf{x}';\omega-i\epsilon) \ . \tag{20b}$$

To second order in the field, we obtain $g_<(1,1')_{t=t'}$ as

$$[g_<(1,1')]_{t=t'}=[g_<^{(0)}(1,1')]_{t=t'}+[g_<^{(1)}(1,1')]_{t=t'}+[g_<^{(2)}(1,1')]_{t=t'} \ , \tag{21}$$

where the linear-response term $g_<^{(1)}(1,1')_{t=t'}$ is given by

$$[g_<^{(1)}(1,1')]_{t=t'}=\int d^42\,G_0^r(1,2)X_{\text{op}}^{(1)}(2)[g_<^{(0)}(2,1')]_{t=t'}+\int d^42\,G_0^{r*}(1',2)X_{\text{op}}^{(1)}(2)[g_<^{(0)}(1,2)]_{t=t'} \ , \tag{22}$$

and the bilinear response (second order) term is

$$\begin{aligned} [g_<^{(2)}(1,1')]_{t=t'}=&\int d^42\,G_0^r(1,2)X^{(2)}(2)[g_<^{(0)}(2,1')]_{t=t'}+\int d^42\,G_0^{r*}(1',2)X^{(2)*}(2)[g_<^{(0)}(1,2)]_{t=t'} \\ &+\int d^42\int d^43\,G_0^r(1,2)X_{\text{op}}^{(1)}(2)G_0^r(2,3)X_{\text{op}}^{(1)}(3)[g_<^{(0)}(3,1')]_{t=t'} \\ &+\int d^42\int d^43\,G_0^{r*}(1',2)X_{\text{op}}^{(1)*}(2)G_0^r(1,3)X_{\text{op}}^{(1)}(3)[g_<^{(0)}(3,2)]_{t=t'} \\ &+\int d^42\int d^43\,G_0^{r*}(1',2)X_{\text{op}}^{(1)*}(2)G_0^{r*}(2,3)X_{\text{op}}^{(1)*}(3)[g_<^{(0)}(1,3)]_{t=t'} \ . \end{aligned} \tag{23}$$

The linear and bilinear current response functions $\langle \mathbf{j}^{(1)} \rangle$ and $\langle \mathbf{j}^{(2)} \rangle$ follow from Eq. (9),

$$\langle \mathbf{j}^{(1)}(1) \rangle = \frac{ie^2}{m} \lim_{\mathbf{x}' \to \mathbf{x}} \mathbf{A}_e(1)[g_<^{(0)}(1,1')]_{t=t'} - \frac{e}{2m} \lim_{\mathbf{x}' \to \mathbf{x}} [\nabla - \nabla' - 2ie\mathbf{a}(\mathbf{x})][g_<^{(1)}(1,1')]_{t=t'} \tag{24}$$

$$\langle \mathbf{j}^{(2)}(1) \rangle = \frac{ie^2}{m} \lim_{\mathbf{x}' \to \mathbf{x}} \mathbf{A}_e(1)[g_<^{(1)}(1,1')]_{t=t'} - \frac{e}{2m} \lim_{\mathbf{x}' \to \mathbf{x}} [\nabla - \nabla' - 2ie\mathbf{a}(\mathbf{x})][g_<^{(2)}(1,1')]_{t=t'} , \tag{25}$$

and the corresponding quantum-transport coefficients readily follow.

The distinctive feature of these current response coefficients is that the time development of the system is fully subsumed in the structure of the retarded Green's function alone for the impurity scattering mechanism under consideration. While our treatment of linear and bilinear response is exact and directly applicable to mesoscopic systems,[15] care must be exercised in averaging over randomized impurity sites. The results above are well adapted to describe steady-state dc- and ac- as well as transient-current response to time-dependent and spatially inhomogeneous electric fields, and can also accommodate an ambient magnetic field as well.

ACKNOWLEDGMENTS

The authors wish to acknowledge helpful discussions with H. C. Tso.

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[1] P. C. Martin and J. Schwinger, Phys. Rev. **115**, 1342 (1959), see Sec. VI for a discussion of the generating Green's function.

[2] N. Ashby, Ph.D. thesis (Harvard University, Cambridge, 1961); N. Ashby, in *Martin-Schwinger Methods in the Many-Body Problem*, Vol. 3 of *Lectures in Theoretical Physics*, edited by W. E. Brittin, B. W. Downs, and J. Downs (Interscience, New York, 1961). In obtaining our Eq. (14) from Ashby's thesis, one should carefully note his comments on pp. 17–20.

[3] L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (Benjamin, Reading, Mass., 1962). The derivation of their Eqs. (8)–(21) provides our Eq. (15).

[4] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1515 (1964) [Sov. Phys.-JETP **20**, 1018 (1965)].

[5] R. A. Craig, J. Math. Phys. **9**, 605 (1968).

[6] D. Langreth, in *Linear and Nonlinear Electron Transport in Solids*, edited by J. T. Devreese and V. E. van Doren (Plenum, New York, 1976).

[7] P. Danielewicz, Ann. Phys. (N.Y.) **152**, 239 (1984); **152**, 305 (1984).

[8] K. Chou, Z. Su, B. Hao, and L. Yu, Phys. Rep. **118**, 1 (1985).

[9] J. Rammer and H. Smith, Rev. Mod. Phys. **58**, 323 (1986).

[10] G. Mahan, Phys. Rep. **145**, 251 (1987).

[11] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957). See also, R. B. Stinchcombe, in *Correlation Functions and Quasiparticle Interactions in Condensed Matter*, Vol. 35 of *NATO Advanced Study Institute, Series B: Physics*, edited by J. Woods (Plenum, New York, 1978), pp. 3–44.

[12] R. L. Peterson, Rev. Mod. Phys. **39**, 69 (1967).

[13] T. Tanaka *et al.*, Phys. Rev. **155**, 388 (1967).

[14] W. Bernard and H. B. Callen, Phys. Rev. **31**, 1017 (1959).

[15] Y. Imry, in Proceedings of the NORDITA Conference on Fundamental Effects in Semiconductor Systems, Neils Bohr Institute, Copenhagen, 1986 (unpublished).