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Reformulation of Steady State Nonequilibrium Quantum Statistical Mechanics

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Starting from the usual formulation of nonequilibrium quantum statistical mechanics, the expectation value of an operator A in a steady state nonequilibrium quantum system is shown to have the form $\langle A\rangle = \mathrm{Tr}\{e^{-\beta(H-Y)}A\}/\mathrm{Tr}\{e^{-\beta(H-Y)}\}$, where H is the Hamiltonian, β is the inverse of the temperature, and Y is an operator which depends on how the system is driven out of equilibrium. Because $\langle A\rangle$ is not expressed as a sum of correlation functions integrated over real time, one can now consider performing *nonperturbative* calculations in *interacting nonequilibrium* quantum problems.

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Nonequilibrium problems have come under increasing study in condensed matter physics. On the one hand, there exists a growing number of classical systems which undergo a phase transition as one drives them out of equilibrium. On the other hand, with technological advances allowing one to make measurements on smaller samples, it becomes easier to drive systems out of equilibrium. This is particularly true in making resistance measurements on very small (mesoscopic) devices at low temperatures [1], which is an inherently *quantum* problem. Furthermore, while linear response measurements do probe some equilibrium correlation functions, much more information can be obtained from the nonlinear response, which probes the full nonequilibrium problem. For example, nonlinear current-voltage characteristics in metal-insulator-superconductor tunnel junctions have long been used to determine the superconducting gap and the phonon density of states. In mesoscopic systems similar kinds of information about the density of states can be obtained from nonlinear transport [2–5].

In spite of the experimental importance of studying nonlinear response there are far fewer theoretical techniques available for studying nonequilibrium quantum systems than for studying equilibrium ones. Again using the example of mesoscopic systems, the noninteracting quantum problem may be solved exactly using the scattering states for electrons coming from reservoirs which are at different chemical potentials. This is the essence of the Landauer formula [6] and its subsequent generalizations [7,8]. For an interacting system the nonlinear current-voltage characteristic can be computed by doing perturbation theory in the part of the Hamiltonian which drives the system out of equilibrium, e.g., an electric field or the tunneling between two leads at different chemical potentials. This perturbation theory, which we will call nonequilibrium quantum statistical mechanics, involves summing a set of real time correlation functions for the linear, quadratic, cubic, etc., response to this interaction [9,10].

While there are many problems for which a perturbation theory is perfectly satisfactory, there is a large class of problems of current interest for which a nonperturbative approach would be quite useful and perhaps even essential. In the case of mesoscopic systems it is now possible to tunnel through a one-dimensional wire [11], a small quantum dot [12], or a single defect [13] in which the electron-electron interaction is large. These systems may exhibit the Kondo effect [14], the fractional quantum Hall effect [15], and perhaps even Luttinger liquid behavior [16]. Nonperturbative calculations, such as renormalization group, Monte Carlo, and variational approaches, have been essential in understanding these phenomena.

The purpose of this paper is to rewrite the conventional perturbation theory of nonequilibrium quantum statistical mechanics for steady state problems in a form similar to that of equilibrium quantum statistical mechanics. This provides an explicit expression for the nonequilibrium density matrix and also the framework upon which one can build nonperturbative calculations in nonequilibrium quantum systems. To be more precise let us define what we mean by the equilibrium and nonequilibrium formulations of quantum statistical mechanics. In equilibrium the expectation value of an operator A is just the trace of A in the Schrödinger picture weighted by the Boltzmann factor $e^{-\beta(H-\mu N)}$, where H is the Hamiltonian, μ is the chemical potential, and N is the number operator:

$$\langle A\rangle = \frac{\mathrm{Tr}\{e^{-\beta(H-\mu N)}A\}}{\mathrm{Tr}\{e^{-\beta(H-\mu N)}\}} . \tag{1}$$

Unless otherwise noted all operators are in the Schrödinger picture.

In the nonequilibrium formulation one only assumes that the system is in equilibrium at some initial time, taken here to be $t=-\infty$. A perturbation H_1 is turned on adiabatically in time, $H=H_0+H_1e^{\eta t}$, to drive the system out of equilibrium. The expectation value of the operator A is its trace in the Heisenberg picture at $t=0$ weighted by the initial distribution function ρ_0 [17]:

$$\langle A\rangle = \frac{\mathrm{Tr}\{\rho_0 A_H(0)\}}{\mathrm{Tr}\{\rho_0\}} . \tag{2}$$

Equation (2) is usually written in the interaction picture, where it is easily expanded in powers of $H_{1I}(t)=e^{iH_0t}H_1$

$\times e^{-iH_0 t} e^{\eta t}$:

$$\langle A\rangle = \frac{1}{\mathrm{Tr}\{\rho_0\}}\mathrm{Tr}\left\{\rho_0\left(A - i\int_{-\infty}^{0} dt_1 [A, H_{1I}(t_1)] + (-i)^2\int_{-\infty}^{0} dt_1 \int_{-\infty}^{t_1} dt_2 [[A, H_{1I}(t_1)], H_{1I}(t_2)] + \cdots\right)\right\}. \tag{3}$$

The term proportional to H_1 is just the familiar linear response of A [18]. The various schemes for doing perturbation theory in nonequilibrium quantum systems, e.g., Kadanoff and Baym [9] or Keldysh [10], are clever ways of summing this series. Although the interaction was turned on adiabatically, in practice one can usually set $\eta = 0$ once the perturbation series is written in the form of Eq. (3) because there is a *physical* relaxation process which causes correlation functions to decay at long times. In this paper we do the real time integrals of Eq. (3) assuming such a relaxation process exists and obtain an expression of the form of Eq. (1) with the operator μN replaced by a new operator Y. The operator $H - Y$ acts as an effective Hamiltonian with which one can develop nonperturbative calculations.

For an equilibrium system the two approaches should give the same result for $\langle A\rangle$. To see this we compare the expansions of Eqs. (1) and (3) in powers of H_1. Clearly, to zeroth order in H_1, they are the same. The linear order term in Eq. (3) can be rewritten using $\mathrm{Tr}\{\rho_0[A, H_{1I}(t_1)]\} = \mathrm{Tr}\{[H_{1I}(t_1), \rho_0]A\}$. Defining a new operator ρ_1, whose time derivative is $\partial\rho_{1I}(t_1)/\partial t_1 = i[\rho_0, H_{1I}(t_1)]$, the real time integral for the linear order contribution to $\langle A\rangle$ can be done explicitly:

$$\begin{aligned}\langle A\rangle_1 &= \frac{1}{\mathrm{Tr}\{\rho_0\}}\int_{-\infty}^{0} dt_1 \mathrm{Tr}\left\{\frac{\partial\rho_{1I}(t_1)}{\partial t_1} A\right\} \\ &= \frac{\mathrm{Tr}\{\rho_{1I}(0)A\}}{\mathrm{Tr}\{\rho_0\}} - \lim_{t\to-\infty}\frac{\mathrm{Tr}\{\rho_{1I}(t)A\}}{\mathrm{Tr}\{\rho_0\}}.\end{aligned} \tag{4}$$

Up to this point we have just made formal manipulations. Now we must make use of the assumption that a physical relaxation process exists and causes the correlation functions to decay in time. The time t in the lower limit of integration is taken to be much larger than this relaxation time so that the integral has converged. Furthermore, because η is a positive infinitesimal we can also take η^{-1} to be much larger than $|t|$, i.e., $\eta|t| \ll 1$. In this limit ρ_{1I} of Eq. (4) becomes the linear order expansion of $e^{-\beta(H-\mu N)}$ in the interaction picture, $\rho_{1I}(t_1) = e^{iH_0 t_1}\rho_1 e^{-iH_0 t_1}$. Finally, we assume that because of the relaxation process the product of the expectation values of two operators, $\mathrm{Tr}\{\rho_0 B_I(t) C_I(t')\}/\mathrm{Tr}\{\rho_0\}$, reduces to the product of their expectation values in the limit that t and t' are separated by much more than the relaxation time:

$$\lim_{|t-t'|\to-\infty}\frac{\mathrm{Tr}\{\rho_0 B_I(t) C_I(t')\}}{\mathrm{Tr}\{\rho_0\}} = \frac{\mathrm{Tr}\{\rho_0 B\}}{\mathrm{Tr}\{\rho_0\}}\frac{\mathrm{Tr}\{\rho_0 C\}}{\mathrm{Tr}\{\rho_0\}}. \tag{5}$$

Making this assumption with $B_I(t) = \rho_0^{-1}\rho_{1I}(t)$ and $C_I(0) = A$, we obtain

$$\langle A\rangle_1 = \frac{\mathrm{Tr}\{\rho_1 A\}}{\mathrm{Tr}\{\rho_0\}} - \frac{\mathrm{Tr}\{\rho_1\}}{\mathrm{Tr}\{\rho_0\}}\frac{\mathrm{Tr}\{\rho_0 A\}}{\mathrm{Tr}\{\rho_0\}}. \tag{6}$$

This is the same result obtained when Eq. (1) is expanded to linear order in H_1. The first term comes from the expansion of the numerator of Eq. (1) and the second term from the expansion of the denominator. Thus, within the relaxation process assumption the two formulations are the same to linear order in the interaction.

One can continue this same procedure for the higher order contributions in H_1 of Eqs. (1) and (3). At each stage the real time integral can be done using

$$\frac{\partial\rho_{n+1,I}}{\partial t} = i[\rho_{n,I}(t), H_{1,I}(t)] \tag{7}$$

and using the relaxation process assumption of Eq. (5). Here, $\rho_{n,I}(t)$ is equal to $e^{iH_0 t}\rho_n e^{-iH_0 t}$ in the limit $\eta \to 0$, and ρ_n is the term in $e^{-\beta(H_0+H_1-\mu N)}$ proportional to $(H_1)^n$. Thus, in equilibrium the two formulations are equivalent when there is a relaxation time in the problem which allows the system to "forget" that it initially had Hamiltonian H_0. Also, note that this proof is not the same as the Gell-Mann and Low theorem [19], which does not involve any assumption about a physical relaxation process.

What happens to this procedure when we go out of equilibrium? First, one must define how the system is driven out of equilibrium. We list here three ways that one can do this for a steady state problem. (i) Initially at $t = -\infty$ one can start with two or more subsystems (reservoirs) at different chemical potentials [20]. Turning on the hopping between the different subsystems drives a current. By assuming that the subsystems are infinitely large, they never equilibrate, and one obtains a steady state nonequilibrium problem. (ii) One can start with an initial density matrix ρ_0 corresponding to a noninteracting nonequilibrium system. Instead of having states in different reservoirs, here one has scattering states at different chemical potentials. Because the scattering states do not interact, the system again consists of subsystems at different chemical potentials. Turning on a many body interaction causes the scattering states to mix, resulting in a true nonequilibrium problem. (iii) One can make an ansatz for the noninteracting nonequilibrium density matrix corresponding to a current source as opposed to a voltage source as in (ii) [8]. Again a many body interaction is turned on adiabatically. In each of these examples the initial density matrix has the form

$$\rho_0 = e^{-\beta(H_0 - Y_0)}, \tag{8}$$

where Y_0 is not μN. For the case of (i) above Y_0 is the sum of the $\mu_i N_i$ for the different subsystems i. The important difference between the equilibrium and the nonequilibrium cases is that Y_0 does not commute with the perturbation H_1. Although it is often convenient to let H_0 be a one body operator, the following derivation is valid for an arbitrary H_0.

There were two key elements in the equilibrium proof: doing the real time integral using the differential equation for ρ_n [Eq. (7)] and taking the limit as the initial time goes to $-\infty$ [Eq. (5)]. The ρ_n are changed because the equilibrium and nonequilibrium distribution functions are different; however, there are still a set of ρ_n which satisfy Eq. (7). Also, in the nonequilibrium case operators still become uncorrelated at long times because of the relaxation process assumption. Thus, the proof remains the same. All we must do is determine the nonequilibrium ρ_n which satisfy Eqs. (7) and (8).

To obtain an expression for the nonequilibrium ρ_n a new set of operators Y_n are introduced. The operator Y_{n+1} is the integral of the commutator of Y_n with H_1:

$$\frac{\partial Y_{n+1,I}(t)}{\partial t}=i[Y_{n,I}(t),H_{1I}(t)]\,. \tag{9}$$

Letting $X_{i,I}(t)=H_{i,I}(t)-Y_{i,I}(t)$ for $i=0,1$ and $X_{i,I}(t)=-Y_{i,I}(t)$ for $i\geq 2$, it is straightforward to verify that the ρ_n for $n\geq 1$ given by

$$\rho_{n,I}(t)=\sum_{m=1}^{\infty}\frac{(-\beta)^m}{m!}\sum_{i_1+\cdots+i_m=n}X_{i_1,I}(t)\cdots X_{i_m,I}(t) \tag{10}$$

satisfy Eqs. (7) and (8). The equilibrium expression for ρ_n has this same form with $X_{0,I}(t)=H_0-\mu N$, $X_{1,I}(t)=H_1$, and $X_{i,I}(t)=0$ for $i\geq 2$. Defining Y to be the sum of the Y_n, $Y=\sum_0^\infty Y_n$, the sum of all the $\rho_n (n\geq 0)$ is $e^{-\beta(H-Y)}$ and the desired expectation value of A out of equilibrium is

$$\langle A\rangle=\frac{\mathrm{Tr}\{e^{-\beta(H-Y)}A\}}{\mathrm{Tr}\{e^{-\beta(H-Y)}\}}\,. \tag{11}$$

This is the principle result of this paper.

What is the operator Y? The differential equation for Y_n, Eq. (9), may be rewritten in terms of commutators as

$$[H_0,Y_n]-i\eta Y_n=[Y_{n-1},H_1]\,, \tag{12}$$

where the positive infinitesimal η has been included to make the operator equation well defined. It is still not important for the convergence of the real time integrals because of the relaxation process assumption. From Eq. (12) and $[H_0,Y_0]=0$ it follows that $[Y,H]=i\eta(Y_0-Y)\to 0$. Thus, the effect of the Y in $e^{-\beta(H-Y)}$ is to change the distribution of the many body eigenstates, not the eigenstates themselves. Because Y commutes with H, we interpret Y as the operator into which Y_0 "evolves" under the perturbation H_1.

This can be made more explicit by considering a simple example. Suppose our initial ($t=-\infty$) system consists of two reservoirs with chemical potentials μ_L and μ_R for left and right [Fig. 1(a)]. Let the operator $c_{k_R}^\dagger$ create a single particle state in the right-hand reservoir and similarly for $c_{k_L}^\dagger$. The operator Y_0 is $\mu_L N_L+\mu_R N_R$, where N_R is the sum $\sum_{k_R} c_{k_R}^\dagger c_{k_R}$ and similarly for N_L. Upon turning on the interaction H_1, which here includes a hopping term between the L and R reservoirs and a many body interaction, a single particle state in the right-hand lead evolves into a scattering state via the Lippmann-Schwinger equation. There is an analogous equation for operators, where the operator $c_{k_R}^\dagger$ is said to evolve into a scattering state operator $\psi_{k_R}^\dagger$. Letting $(\psi_{k_R}^\dagger)_n$ be the contribution to $\psi_{k_R}^\dagger$ proportional to $(H_1)^n$, this equation reads ($n\geq 1$)

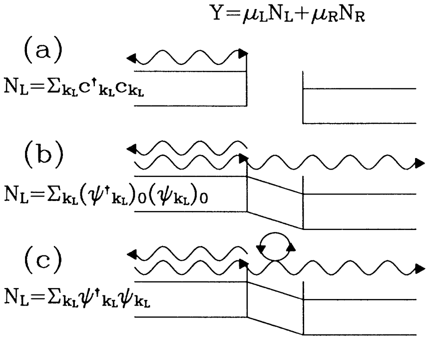

FIG. 1. Schematic representation for the operator Y, which appears in the nonequilibrium density matrix $e^{-\beta(H-Y)}$. In this example there are two reservoirs with potential offsets and different chemical potentials. For all the cases Y is equal to $\mu_L N_L+\mu_R N_R$. (a) For the disconnected system, N_L and N_R just measure the number of particles on the left and right. (b) For the connected noninteracting system, N_L measures the number of noninteracting scattering states moving from the left to the right. (c) For the connected interacting system, N_L is expressed in terms of many body scattering operators ψ_{k_L}, which include particle-hole excitations (depicted as a circle) and other many body excitations.

$$\frac{d(\psi_{k_R}^\dagger)_{n,I}}{dt}-i\epsilon_{k_R}(\psi_{k_R}^\dagger)_{n,I}=i[(\psi_{k_R}^\dagger)_{n-1,I},H_1]\,, \tag{13}$$

where $(\psi_{k_R}^\dagger)_0=c_{k_R}^\dagger$. It is important to note that for the case of noninteracting electrons this operator creates a conventional scattering state [Fig. 1(b)], while for the full interacting problem it is a full many body operator consisting of particle-hole excitations, etc. [Fig. 1(c)]. Using Eq. (13) and the similar equations for the left-hand scattering states, it follows that Y_n defined as the contribution proportional to $(H_1)^n$ in

$$Y=\mu_R\sum_{k_R}\psi_{k_R}^\dagger\psi_{k_R}+\mu_L\sum_{k_L}\psi_{k_L}^\dagger\psi_{k_L} \tag{14}$$

satisfies Eq. (9). Thus, we have been able to interpret the derived result for Y in terms of many body scattering operators.

The conventional tunneling formalism or Landauer formula is modified by including many body scattering *operators* in the exponential instead of single particle ones. It is important to note that putting scattering operators in the exponential does not mean that the density matrix can be expressed as a Slater determinant of scattering states as in the noninteracting case. Also, although H_0 in this example is quadratic (noninteracting), the derivation of Y presented earlier is true for an arbitrary H_0.

All of the above discussion deals with the expectation value of a single operator A. To compute the expectation value of two operators at different times, e.g., $\langle A(t) \times B(t')\rangle$, a similar derivation shows that the time dependence is included via H: $A(t)=e^{iHt}Ae^{-iHt}$. This illustrates one of the principal differences between equilibrium and nonequilibrium problems: The operator which governs the time dependence, H, is not the same as the operator which governs the occupation factors, $H-Y$. This is the reason that the fluctuation dissipation theorem breaks down as one goes out of equilibrium. Recall that the key relation which leads to the fluctuation dissipation theorem [21] in equilibrium is that $\langle A(t-i\beta)B(t')\rangle$ is equal to $\langle B(t')A(t)\rangle$. For this class of nonequilibrium problems one finds instead that

$$\langle A(t-i\beta)B(t')\rangle=\langle e^{-\beta Y}B(t')e^{\beta Y}A(t)\rangle . \tag{15}$$

The additional contributions besides $\langle B(t')A(t)\rangle$ are computed by expanding $e^{-\beta Y}B(t')e^{\beta Y}$ in Y.

In this paper we have reformulated traditional nonequilibrium quantum statistical mechanics in a way which does not involve response functions in real time. This answers important conceptual questions: What is the nonequilibrium density matrix? How does the fluctuation dissipation theorem break down? It also opens the door to doing nonperturbative calculations in nonequilibrium quantum problems. For example, one can minimize the operator $H-Y$ to obtain the steady state solution to a nonequilibrium problem at low temperatures.

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