Physica A 164 (1990) 469–484
North-Holland

STEADY STATE OF PERIODICALLY DRIVEN SYSTEMS IN QUANTUM STATISTICS

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Received 27 September 1989

A formal theory for the statistical operator of the "steady state" in periodically driven matter is given. For this purpose the periodic fields are changed to have an amplitude increasing with $e^{\eta t}$, where η is taken to be zero at the end. This procedure allows for a correct description of energy denominators. The results following from the steady state operator are compared with the long time behaviour obtained from nth order response and are shown to agree. Thus the proposed statistical operator has all properties required.

1. Introduction

A quantum statistical treatment of perodically driven macroscopic matter can be based on response theory or equations of motion for expectation values. They may be derived from the von Neumann equation with some approximations. In both cases the time dependence of the expectation values shows some relaxation behaviour before a periodic motion is finally reached[#1]. The most simple example in this respect is given by the Bloch equations with a rotating transverse field. The steady state can be studied by such equations and thus is limited to the range of validity of the equations. Therefore the question arises if it is possible to find a statistical operator for the "steady state" directly from the von Neumann equation, without a microscopic derivation of equations for expectation values. A first attempt in this direction was done by some authors [1] who proposed a Floquet decomposition of the time evolution operator $U(t)$,

$$U(t) = \mathrm{T} \exp \int_0^t -\frac{\mathrm{i}}{\hbar}\, \mathcal{H}(t')\, \mathrm{d}t' = Q(t)\, \mathrm{e}^{-(i/\hbar)\bar{\mathcal{H}} t}\,, \tag{1.1}$$

[#1] We do not deal here with the possibility of subharmonics and chaotic motion.

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with periodic $Q(t)$,

$$Q(t+\tau)=Q(t)\,. \tag{1.2}$$

They wanted to deduce the long time behaviour of the statistical operator from this decomposition. Although it is possible to apply eq. (1.1) to simple systems without internal interactions and to obtain expansions [1, 2] of $\tilde{\mathcal{H}}$ for driving frequencies larger than internal resonances, the principal idea could not be carried further. In applying it to many-body systems severe difficulties appear. In our opinion there are two reasons which are connected.

(i) As already discussed by Maricq [1], the decomposition (1) is not unique. Having found a particular $\tilde{\mathcal{H}}=\sum E_n P_n$ one can change the eigenvalues E_n by $2\pi m_n \tau^{-1}$ with integer m_n and define a new operator

$$\bar{\mathcal{H}}=\sum_n (E_n+2\pi m_n \tau^{-1})P_n\,, \tag{1.3}$$

so that

$$U=\bar{Q}(t)\,e^{-(i/\hbar)\bar{\mathcal{H}}t}\,, \tag{1.4}$$

with

$$\bar{Q}(t+\tau)=\bar{Q}(t)\,, \tag{1.5}$$

holds.

(ii) From the simple example of harmonic oscillators [2] one can see that in general there is no $\tilde{\mathcal{H}}$ which is continuous in the driving frequency $\omega\equiv 2\pi\tau^{-1}$ or the internal energy differences. Passing through resonances one must change the eigenvalues E_n according to eq. (1.3). From this it follows that traces of the form

$$\mathrm{Tr}(F\,e^{-\beta\tilde{\mathcal{H}}}) \tag{1.6}$$

are not well defined in the thermodynamic limit. Enlarging the particle number one passes through resonances with ω.

In this paper we want to show that it is possible to circumvent the difficulties mentioned above by using a driving field $h(t)$ which has a periodic factor $f(t)$:

$$f(t+\tau)=f(t)\,, \tag{1.7}$$

and an increasing amplitude[#2]

$$h(t)=\mathrm{e}^{\eta t}hf(t)\,, \qquad \eta>0\,. \tag{1.8}$$

In this way, taking the limit $\eta \to 0$ at the end, the resonances are treated appropriately. The idea behind this can be easily seen from the case of linear response. Let the linear response function for an observable F be $\varphi(t)$, then for a driving field

$$h(t)=\mathrm{e}^{\eta t}h\,\mathrm{Re}\,\mathrm{e}^{-\mathrm{i}\omega t}\,, \qquad t>0\,, \tag{1.9}$$

one asymptotically obtains the response

$$\langle F\rangle(t)=h\,\mathrm{e}^{\eta t}\,\mathrm{Re}\left(\mathrm{e}^{-\mathrm{i}\omega t}\int_0^\infty \varphi(t')\,\mathrm{e}^{\mathrm{i}\omega t'-\eta t'}\,\mathrm{d}t'\right), \qquad t\gg\tau_{\mathrm{R}}\,. \tag{1.10}$$

For finite particle number the factor $\mathrm{e}^{-\eta t'}$ guarantees that the integral exists and no divergent resonances occur.

Our general treatment of the fields (1.8) is based on section 2, where we establish a decomposition of the time evolution operator $U(t)$ which is similar to the Floquet theorem (1.1) for periodic fields, but leads to a factor $\mathrm{e}^{-(\mathrm{i}/\hbar)\hat{\mathscr{H}}t}$ with unique $\hat{\mathscr{H}}$. In section 3 we then are ready to give the long time behaviour of the statistical operator by making the replacement

$$\mathrm{e}^{-(\mathrm{i}/\hbar)\hat{\mathscr{H}}t}\,\rho(0)\,\mathrm{e}^{(\mathrm{i}/\hbar)\hat{\mathscr{H}}t}\to\frac{\mathrm{e}^{-\beta\hat{\mathscr{H}}}}{Z}\,, \qquad t\gg\tau_{\mathrm{R}}\,. \tag{1.11}$$

This procedure is confirmed in section 4 where we compare the expansion of the asymptotic state resulting from eq. (1.11) with the long time behaviour obtained from response theory for the case that a heat bath is included in the system.

2. Decomposition of the time evolution operator

Let a system be driven by a c-number field of the type (1.8). We regard the Hamiltonian as a function of t and h,

$$\mathscr{H}(h(t))=\mathscr{H}(t,h)\,. \tag{2.1}$$

[#2] One may think of η to have the order of magnitude of the inverse recurrence time.

Then from eq. (1.8) it follows that

$$\mathcal{H}(t+\tau, h) = \mathcal{H}(t, \mathrm{e}^{\eta\tau} h) \tag{2.2}$$

holds. We now want to show that eq. (2.2) implies that the time evolution operator $U(t, h)$ resulting from

$$\frac{\mathrm{d}U}{\mathrm{d}t} = -\frac{\mathrm{i}}{\hbar}\, \mathcal{H}(t, h)\, U\,, \qquad U = \mathbb{1} \quad \text{for } t = 0\,, \tag{2.3}$$

can be decomposed in the following way[#3]:

$$U(t, h) = Q(t, h)\, C(\mathrm{e}^{\eta t} h)^{\dagger}\, C(h)\, \mathrm{e}^{(\mathrm{i}/\hbar) C(h)^{\dagger} \mathcal{H} C(h) t}\,, \tag{2.4}$$

where

$$Q(t+\tau, h) = Q(t, \mathrm{e}^{\eta\tau} h)\,. \tag{2.5}$$

Here $\mathcal{H}$ denotes the Hamiltonian with no driving field,

$$\mathcal{H} = \mathcal{H}(h(t) = 0)\,, \tag{2.6}$$

and C depending on h, η, ω is defined by

$$C(h) = \lim_{t \to -\infty} \mathrm{e}^{(\mathrm{i}/\hbar)\mathcal{H} t}\, U(t, h)\,. \tag{2.7}$$

To derive eq. (2.4) we take into account the dependence of U on the parameter h explicitly, $U = U(t, h)$. Then from eqs. (2.3) and (2.2) we find

$$\frac{\mathrm{d}}{\mathrm{d}t}\, U(t+\tau, h) = -\frac{\mathrm{i}}{\hbar}\, \mathcal{H}(t, \mathrm{e}^{\eta\tau} h)\, U(t+\tau, h)\,. \tag{2.8}$$

This equation can be integrated with the help of $U(t, \mathrm{e}^{\eta\tau} h)$. Thus we have the relationship

$$U(t+\tau, h) = U(t, \mathrm{e}^{\eta\tau} h)\, U(\tau, h)\,. \tag{2.9}$$

On the other hand, for $t \to -\infty$ the field vanishes,

$$\lim_{t \to -\infty} \mathcal{H}(t, h) = \mathcal{H}\,, \tag{2.10}$$

[#3] $C^{\dagger} \mathcal{H} C = \tilde{\mathcal{H}}$ mentioned in the introduction.

so that one can integrate eq. (2.3) to give

$$U(t, h) = \mathrm{e}^{-(\mathrm{i}/\hbar)\mathscr{H}t} C(h) \qquad \text{for } t \to -\infty\,, \tag{2.11}$$

which is equivalent to eq. (2.7). Regarding eq. (2.9) formally for $t \to -\infty$ and inserting the expressions (2.11) we find

$$\mathrm{e}^{-(\mathrm{i}/\hbar)\mathscr{H}(t+\tau)} C(h) = \mathrm{e}^{-(\mathrm{i}/\hbar)\mathscr{H}t} C(\mathrm{e}^{\eta\tau} h)\, U(\tau, h)\,, \tag{2.12}$$

or

$$C(\mathrm{e}^{\eta\tau} h)^{\dagger}\, \mathrm{e}^{-(\mathrm{i}/\hbar)\mathscr{H}\tau}\, C(h) = U(\tau, h)\,. \tag{2.13}$$

Define $Q(t, h)$ by

$$Q(t, h) = U(t, h) C(h)^{\dagger}\, \mathrm{e}^{-(\mathrm{i}/\hbar)\mathscr{H}t}\, C(\mathrm{e}^{\eta t} h)\,; \tag{2.14}$$

then using eq. (2.9) and inserting $U(\tau, h)$ from eq. (2.13) we find the property

$$\begin{aligned} Q(t+\tau, h) &= U(t, \mathrm{e}^{\eta\tau} h)\, C(\mathrm{e}^{\eta\tau} h)^{\dagger}\, \mathrm{e}^{-(\mathrm{i}/\hbar)\mathscr{H}\tau} C(h)\, C(h)^{\dagger}\, \mathrm{e}^{(\mathrm{i}/\hbar)\mathscr{H}(t+\tau)} C(\mathrm{e}^{\eta(t+\tau)} h) \\ &= U(t, \mathrm{e}^{\eta\tau} h)\, C(\mathrm{e}^{\eta\tau} h)^{\dagger}\, \mathrm{e}^{(\mathrm{i}/\hbar)\mathscr{H}t} C(\mathrm{e}^{\eta t}\, \mathrm{e}^{\eta\tau} h) = Q(t, \mathrm{e}^{\eta\tau} h)\,, \end{aligned} \tag{2.15}$$

which together with eq. (2.14) proves the decomposition (2.4).

The basic relation (2.4) is similar to the Floquet theorem. The operator $Q(t, h)$ has the same property as the Hamiltonian (2.2). If we take η so small that for all physical times $\mathrm{e}^{\eta t} \approx 1$ we find that, within times $t \ll \eta^{-1}$, $Q(t, h)$ is periodic and eq. (2.4) reduces to a decomposition of the Floquet type which, however, is different from the case $\eta = 0$, since $Q(t, h)$ and $C(h)$ still depend on $\eta \neq 0$. Nevertheless one should bear in mind that, having calculated all traces, in final results functions of the form

$$\frac{1}{\eta}(\mathrm{e}^{\eta t} - 1) \to t \tag{2.16}$$

may occur. This is the reason why we prefer to keep the time dependence with $\mathrm{e}^{\eta t}$ up to the end.

3. The steady state

We want to calculate expectation values of macroscopic observables in the long time limit, i.e. for times larger than internal relaxation times, using the

Schrödinger representation. We start at $t=0$ with the statistical operator $\rho(0)$. Then the exact statistical operator at time t follows from eq. (2.4) to be[#4]

$$\rho(t) = U(t,h)\,\rho(0)\,U(t,h)^{\dagger} \tag{3.1}$$

$$= Q(t)\,C(t)^{\dagger}\,C\,\mathrm{e}^{-(\mathrm{i}/\hbar)C^{\dagger}\mathcal{H}Ct}\,\rho(0)\,\mathrm{e}^{(\mathrm{i}/\hbar)C^{\dagger}\mathcal{H}Ct}\,C^{\dagger}C(t)\,Q(t)^{\dagger}\,. \tag{3.2}$$

It consists of a unitary transformation which is approximately periodic and a time dependence known from relaxation processes. We assume that the undriven system described by $\mathcal{H}$ reaches equilibrium after a relaxation time τ_{R}, then it is appealing to take for long times in eq. (3.2):

$$\mathrm{e}^{-(\mathrm{i}/\hbar)C^{-1}\mathcal{H}Ct}\rho(0)\,\mathrm{e}^{(\mathrm{i}/\hbar)C^{-1}\mathcal{H}Ct} \to \frac{\mathrm{e}^{-\beta C^{-1}\mathcal{H}C}}{Z}\,, \qquad t \gg \tau_{\mathrm{R}}\,, \tag{3.3}$$

where the parameter β will be fixed later. This means that for times longer than internal relaxation times τ_{R} it should hold that

$$\langle F\rangle(t) = \mathrm{Tr}[F\rho(t)] = \mathrm{Tr}[F\rho_{\mathrm{st}}(t)] \tag{3.4}$$

with the operator of the steady state given by

$$\rho_{\mathrm{st}}(t) = Q(t)\,C(t)^{\dagger}\,\frac{\mathrm{e}^{-\beta\mathcal{H}}}{Z}\,C(t)\,Q(t)^{\dagger} = \frac{1}{Z}\,\mathrm{e}^{-\beta Q(t)\,C(t)^{\dagger}\,\mathcal{H}C(t)\,Q(t)^{\dagger}}\,. \tag{3.5}$$

Let us study the replacement of $\rho(t)$ by $\rho_{\mathrm{st}}(t)$ in more detail. The expectation value of an observable F with $\rho(t)$ (3.1) can be written in the following form:

$$\langle F\rangle(t) = \mathrm{Tr}[F\rho(t)]$$

$$= \mathrm{Tr}(C(t)\,Q(t)^{\dagger}\,FQ(t)\,C(t)^{\dagger}\,\mathrm{e}^{-(\mathrm{i}/\hbar)\mathcal{H}t}C\rho(0)\,C^{\dagger}\,\mathrm{e}^{(\mathrm{i}/\hbar)\mathcal{H}t})\,, \tag{3.6}$$

where use has been made of the cyclic invariance of the trace. Eq. (3.6) may be read as a calculation in a special interaction representation with

$$F(t,h) = C(\mathrm{e}^{\eta t}h)\,Q(t,h)^{\dagger}\,FQ(t,h)\,C(\mathrm{e}^{\eta t}h)^{\dagger}\,. \tag{3.7}$$

To find the long time behaviour in eq. (3.6) we will expand $F(t,h)$. From the property (2.5) of $Q(t,h)$ it follows that

$$F(t,\mathrm{e}^{-\eta t}h) = F(t+\tau,\mathrm{e}^{-\eta(t+\tau)}h) \tag{3.8}$$

[#4] For brevity we denote $C = C(h)$; $C(t) = C(\mathrm{e}^{\eta t}h)$; $Q(t) = Q(t,h)$.

is periodic with τ, and thus can be represented as a Fourier series. Hence, replacing h by $e^{\eta t}h$, we can write

$$F(t, h) = \sum_m e^{im\omega t} F_m(h\, e^{\eta t}) . \tag{3.9}$$

For the expectation value (3.6) we therefore get

$$\langle F\rangle(t) = \sum_m e^{im\omega t}\, \mathrm{Tr}[F_m(h\, e^{\eta t})\, e^{-(i/\hbar)\mathscr{H}t}\, C\rho(0)\, C^\dagger\, e^{(i/\hbar)\mathscr{H}t}] . \tag{3.10}$$

Assume uniform convergence of the factors defined by the traces for fixed fields h_1 in the range

$$h < h_1 < h\, e^{\eta t} ; \tag{3.11}$$

then the asymptotic behaviour for $\tau_R \ll t < t_1$ is given by

$$\mathrm{Tr}[F_m(h\, e^{\eta t})\, e^{-(i/\hbar)\mathscr{H}t}\, C\rho(0)\, C^\dagger\, e^{(i/\hbar)\mathscr{H}t}] \to \mathrm{Tr}\left(F_m(h\, e^{\eta t})\, \frac{e^{-\beta\mathscr{H}}}{Z}\right) . \tag{3.12}$$

The condition (3.11) is very weak, since for all physically admissible times $t < t_1$ we can choose η to be so small that $e^{\eta t_1} \approx 1$ holds. Inserting eq. (3.12) into eq. (3.10) and using eq. (3.9) we arrive at the results (3.3) and (3.4).

We are left with the question, how to fix the parameter β in $\rho_{st}(t)$. Here an arguing similar to thermodynamics is possible. Since we can write $\rho_{st}(t)$ as an exponential

$$\rho_{st}(t) = \frac{1}{Z}\, e^{-\beta Q(t)\, C(t)^\dagger\, \mathscr{H} C(t)\, Q(t)^\dagger} , \tag{3.13}$$

the transition from $\rho(t)$ to $\rho_{st}(t)$ means to introduce a generalized canonical statistical operator for which Jaynes' concept of maximum uncertainty applies [3]. Therefore β is to be determined in such a way that the expectation values of $Q(t)\, C(t)^\dagger\, \mathscr{H} C(t)\, Q(t)^\dagger$, taken with $\rho(t)$ and $\rho_{st}(t)$, agree for $\tau_R \ll t$:

$$\mathrm{Tr}[Q(t)\, C(t)^\dagger\, \mathscr{H} C(t)\, Q(t)^\dagger\, \rho(t)] = \mathrm{Tr}[Q(t)\, C(t)^\dagger\, \mathscr{H} C(t)\, Q(t)^\dagger\, \rho_{st}(t)] ,$$
$$\tau_R \ll t . \tag{3.14}$$

Inserting the statistical operators (3.1) and (3.5) into this equation we are led to a condition on β:

$$\mathrm{Tr}[\mathscr{H} C\rho(0)\, C^\dagger] = \mathrm{Tr}\left(\mathscr{H}\, \frac{e^{-\beta\mathscr{H}}}{Z}\right) . \tag{3.15}$$

In the case that we start from equilibrium,

$$\rho(0) = \frac{e^{-\beta_i \mathcal{H}}}{Z}\,; \tag{3.16}$$

then eq. (3.15) yields

$$\mathrm{Tr}\left(C^{+}\mathcal{H}C\,\frac{e^{-\beta_i \mathcal{H}}}{Z(\beta_i)}\right) = \mathrm{Tr}\left(\mathcal{H}\,\frac{e^{-\beta \mathcal{H}}}{Z(\beta)}\right). \tag{3.17}$$

This means that β depends on the initial temperature β_i and on the parameters of the driving field,

$$\beta = \beta(\beta_i, h, \omega, \eta)\,. \tag{3.18}$$

If the system described by $\mathcal{H}$ comprises a heat bath and the fields $h(t)$ act on a subsystem, then it is possible to show that due to the large heat capacity of the bath, eq. (3.17) leads to (appendix A)

$$\beta = \beta_i\,. \tag{3.19}$$

4. Comparison with response theory

In this section we will confirm the transition from $\rho(t)$ to $\rho_{st}(t)$ at long times from a different point of view. We want to show that in expanding the expectation values $\mathrm{Tr}[F\rho_{st}(t)]$ into powers of the driving amplitude, the results in each order coincide with those of response theory obtained for long times. In this comparison we restrict to the case that the Hamiltonian comprises a heat bath and the fields act upon a subsystem. The reason for this restriction is that in the general case the long time behaviour of the nth order response is not known yet, since the nth order dynamic susceptibilities may have poles at some frequencies. Only for $n = 2$ the general problem was investigated [4].

We start with the expansion of $\rho_{st}(t, h)$ in eq. (3.5) into powers of h. From eq. (3.19) we have $\beta = \beta_i$, so that we need not take into account the expansions of β. The dependence of $\rho_{st}(t, h)$ on h is caused just by $Q(t, h)$ and $C(e^{\eta t}h)$. Instead of expanding these quantities separately and collecting the n-th order term we expand $\rho_{st}(t, h)$ as a whole. This can be easily achieved, since from eqs. (2.4) and (3.5) it follows that $\rho_{st}(t, h)$ is a solution to the von Neumann equation for which according to eq. (2.10) the extrapolated initial value

$$\rho_{st}(-\infty) = \frac{e^{-\beta\mathcal{H}}}{Z}\ \rho_\beta \tag{4.1}$$

holds. Therefore we can go the usual way of perturbation theory setting up an integral equation for $\rho_{st}(t)$ and iterating it. Let the field be linearly coupled in $\mathcal{H}(t, h)$,

$$\mathcal{H}(t, h) = \mathcal{H} + \mathcal{H}^1(t, h)\,, \tag{4.2}$$

with

$$\mathcal{H}^1(t, h) = e^{\eta\tau} h \sum_m \mathcal{H}_m\, e^{im\omega t}\,, \tag{4.3}$$

and the corresponding Liouvillians be

$$Lx = (1/\hbar)[\mathcal{H}, x]\,, \qquad L^1(t, h)\, x = h\, e^{\eta t} \sum_m (1/\hbar)[\mathcal{H}_m, x]\, e^{im\omega t}\,; \tag{4.4}$$

then from the von Neumann equation

$$\frac{d}{dt}\, \rho_{st}(t, h) = iL\rho_{st}(t, h) + iL^1(t, h)\, \rho_{st}(t, h)\,, \tag{4.5}$$

with the initial condition (4.1), we get the integral equation

$$\rho_{st}(t, h) = \rho_\beta - i \int_{-\infty}^{t} e^{-iL(t-t')} L^1(t', h)\, \rho_{st}(t', h)\, dt'\,, \tag{4.6}$$

from which we find the nth order contribution of $\rho_{st}(t, h)$ to be

$$\rho_{st}^{(n)}(t, h) = (-i)^n \bigg(\int_{-\infty}^{t} e^{-iL(t-t_n)} L^1(t_n)\, dt_n \int_{-\infty}^{t_n} e^{-iL(t_n - t_{n-1})} L^1(t_{n-1})\, dt_{n-1} \cdots$$
$$\cdots \int_{-\infty}^{t_2} e^{-iL(t_2 - t_1)} L^1(t_1)\, dt_1 \bigg) \rho_\beta\,. \tag{4.7}$$

Inserting the expression for $L^1(t, h)$ (4.4) and carrying out the integrations we obtain the result for the nth order contribution of an expectation value $\mathrm{Tr}[F\rho_{st}(t, h)]$ as

$$\mathrm{Tr}[F\rho_{st}(t,h)]^{(n)} = (-1^n)(e^{\eta t}h)^n \sum_{m_1\ldots m_n} e^{i(m_1+m_2+\cdots+m_n)\omega t}$$
$$\times \mathrm{Tr}\Big(F \frac{1}{L+(m_1+m_2+\cdots+m_n)\omega - in\eta}$$
$$\times L_{m_n} \frac{1}{L+(m_1+\cdots+m_{n-1})\omega - i(n-1)\eta} L_{m_{n-1}} \cdots$$
$$\cdots \frac{1}{L+m_1\omega - i\eta} L_{m_1}\rho_\beta\Big). \tag{4.8}$$

It consists of the products of an amplitude $(e^{\eta t}h)^n$ and a periodic function in which the traces are given by nth order dynamic susceptibilities.

The result (4.8) is to be compared with response theory for long times. Here we have to start from the von Neumann equation (4.5) for $\rho(t, h)$ but with the initial condition at $t=0$:

$$\rho(0) = \rho_{\beta_i} = \rho_\beta . \tag{4.9}$$

The corresponding integral equation for $\rho(t, h)$ therefore reads

$$\rho(t,h) = \rho_\beta - i\int_0^t e^{-iL(t-t')} L^1(t',h)\,\rho(t',h)\,dt'$$
$$= \rho_\beta - i\int_0^t e^{-iLt'} L^1(t-t',h)\,\rho(t-t',h)\,dt . \tag{4.10}$$

Iterating it we obtain the nth order contribution to $\rho(t, h)$

$$\rho(t,h)^{(n)} = (-i)^n\Big(\int_0^t e^{-iLt_n} L^1(t-t_n)\,dt_n \int_0^{t-t_n} e^{-iLt_{n-1}}$$
$$\times L^1(t-t_n-t_{n-1})\,dt_{n-1}\cdots$$
$$\cdots \int_0^{t-t_n-t_{n-1}-\cdots-t_2} e^{-iLt_1} L^1(t-t_n-t_{n-1}-\ldots-t_1)\,dt_1\Big)\rho_\beta , \tag{4.11}$$

from which we get the nth order response of $\mathrm{Tr}[F\rho(t,h)]$ to be

$$\mathrm{Tr}[F\rho(t,h)]^{(n)} = (e^{\eta t}h)^n \sum_{m_1\ldots m_n} e^{i(m_1+m_2+\cdots+m_n)\omega t} c_{m_1\ldots m_n}(t) , \tag{4.12}$$

where the time dependent coefficients $c_{m_1\dots m_n}(t)$ are defined by the traces

$$c_{m_1\dots m_n}(t) = (-\mathrm{i})^n \operatorname{Tr}\Big[F\Big(\int_0^t \mathrm{e}^{-\mathrm{i}Lt_n} L_{m_n} \mathrm{e}^{-\mathrm{i}(m_n+m_{n-1}+\dots+m_1)\omega t_n - n\eta t_n}\,\mathrm{d}t_n \cdots$$
$$\cdots \int_0^{t-t_n-t_{n-1}-\dots-t_2} \mathrm{e}^{-\mathrm{i}Lt_1} L_{m_1} \mathrm{e}^{-\mathrm{i}m_1 t_1 - \eta t_1}\,\mathrm{d}t_1\Big)\rho_\beta\Big]\,. \tag{4.13}$$

The long time behaviour of the response (4.12) takes on the form

$$\operatorname{Tr}[F\rho(t,h)]^{(n)} = (\mathrm{e}^{\eta t}h)^n \sum_{m_1\dots m_n} \mathrm{e}^{\mathrm{i}(m_1+\dots+m_n)\omega t} c_{m_1\dots m_n}(\infty)\,, \tag{4.14}$$

since the limits $c_{m_1\dots m_n}(\infty)$ exist.

Let F be an observable of the subsystem of interest: then from the conditions imposed on the total system we conclude that the functions $c_{m_1\dots m_n}(t)$ reach their steady value for times $t \gg \tau_{\mathrm{R}}$ [#5] (appendix B). Evaluating the expressions $c_{m_1\dots m_n}(\infty)$ and inserting them into eq. (4.14) one sees that eq. (4.14) is identical to eq. (4.8). Thus we have shown that for the considered systems

$$[F\rho_{\mathrm{st}}(t,h)]^{(n)} = \operatorname{Tr}[F\rho(t,h)]^{(n)}\,, \qquad t \gg \tau_{\mathrm{R}}\,. \tag{4.15}$$

After having performed the thermodynamic limit one can also take the limit $\eta \to 0$. Then the nth order contribution according to eq. (4.8) or (4.15) is purely periodic.

In the general case without our restrictions imposed on the system and the observables F one can also compare the results for $\operatorname{Tr}[F\rho_{\mathrm{st}}(t,h)]^{(n)}$ for $n=1$ and 2 with those of response theory. One has to expand $\beta(\beta_{\mathrm{i}}, h, \omega, \eta)$. For $n=1$ no problems arise since one finds that in the expansion of β,

$$\beta = \beta_{\mathrm{i}} + \beta^{(1)} + \beta^{(2)} + \cdots\,, \tag{4.16}$$

$$\beta^{(1)} = 0 \tag{4.17}$$

holds, so that eq. (4.8) for $n=1$ is generally valid and coincides with eq. (4.15) without further assumptions. For $n=2$ we find from eq. (4.8):

[#5] If the integrals in eq. (4.13) do not decay in all time arguments $t_1, \dots, t_n$ the asymptotic value of $c_{m_1\dots m_n}(t)$ is not reached before $t \gg \eta^{-1}$. This is physically useless, since we are interested in times $\eta t \ll 1$.

$$\mathrm{Tr}[F\rho_{\mathrm{st}}(t,h)]^{(2)} = (\mathrm{e}^{\eta t}h)^2 \sum_{m_1 m_2} \mathrm{e}^{\mathrm{i}(m_1 - m_2)\omega t}$$

$$\times \mathrm{Tr}\left(F \frac{1}{L + (m_1 + m_2)\omega - 2\mathrm{i}\eta} L_{m_2} \frac{1}{L + m_1\omega - \mathrm{i}\eta} L_{m_1}\right)\rho_{\beta_i}$$

$$+ \beta^{(2)} \frac{\partial}{\partial \beta_i} \mathrm{Tr}(F\rho_{\beta_i}) . \tag{4.18}$$

It is not difficult to realise from eq. (3.17) that $\beta^{(2)}$ contains a term singular in η, which combines with the singular part of the first term in eq. (4.18) to yield a contribution

$$\tfrac{1}{2\eta}(\mathrm{e}^{2\eta t} - 1) \to t . \tag{4.19}$$

This is especially true for choosing $F = \mathcal{H}$ in calculating energy absorption. Evaluating $\beta^{(2)}$ in detail one sees that eq. (4.18) agrees with the general result obtained from second order response [4].

Summarizing, we may state that the steady state operator $\rho_{\mathrm{st}}(t)$ in eq. (3.5) is consistent with all results hitherto known from response theory.

5. Conclusion

We have given a formal prescription how to construct a steady state operator $\rho_{\mathrm{st}}(t)$ for systems which are driven by periodic fields with an increasing amplitude $\mathrm{e}^{\eta t}h$. After the calculation of all traces in the thermodynamic limit, the limit $\eta \to 0$ can be done. The procedure with $\mathrm{e}^{\eta t}$ was necessary for getting a correct description of energy denominators. The results for expectation values $\lim_{\eta\to 0} \mathrm{Tr}[F\rho_{\mathrm{st}}(t)]$ may be either purely periodic or not depending on the system and observables.

The idea behind the formal concept is to be able to study the dependence of $\rho_{\mathrm{st}}(t)$ on the parameters of the driving field similar to the framework of thermodynamics. Perhaps in this way it will be possible to get a better understanding of the thresholds for energy absorption, known to occur in states far from equilibrium.

Appendix A

Evaluation of $\beta = \beta_i$

The parameter β is to be determined from eq. (3.15)

$$\mathrm{Tr}[\mathcal{H}\rho(\beta)] = \mathrm{Tr}[C^{+}\mathcal{H}C\rho(\beta_i)] . \tag{A.1}$$

Let the system be composed of a subsystem (S) coupled to a bath (B). The driving fields act on the subsystem only, hence

$$\mathcal{H}(t,h)=\mathcal{H}+\mathcal{H}_{\mathrm{S}}^{1}(t,h)\,. \tag{A.2}$$

First, we note that instead of calculating the double limits for

$$\mathrm{Tr}[C^{\dagger}\mathcal{H}C\rho(\beta_{\mathrm{i}})]=\lim_{t_1\to-\infty,t_2\to-\infty}\mathrm{Tr}[U(t_1)^{\dagger}\,\mathrm{e}^{(\mathrm{i}/\hbar)\mathcal{H}t_1}\,\mathcal{H}\,\mathrm{e}^{-(\mathrm{i}/\hbar)\mathcal{H}t_2}U(t_2)\rho(\beta_{\mathrm{i}})]\,, \tag{A.3}$$

we may also write

$$\mathrm{Tr}[C^{\dagger}\mathcal{H}C\rho(\beta_{\mathrm{i}})]=\lim_{t\to-\infty}\mathrm{Tr}[U(t)^{\dagger}\mathcal{H}U(t)\;\rho(\beta_{\mathrm{i}})]\,, \tag{A.4}$$

since from the long time behaviour of $U(t)$,

$$U(t)=\mathrm{e}^{-(\mathrm{i}/\hbar)\mathcal{H}t}C\,,\qquad t\to-\infty\,, \tag{A.5}$$

it follows that

$$\begin{aligned}\mathrm{Tr}[U(t)^{\dagger}\;\mathcal{H}U(t)\;\rho(\beta_{\mathrm{i}})]&=\mathrm{Tr}[C^{\dagger}\,\mathrm{e}^{(\mathrm{i}/\hbar)\mathcal{H}t}\,\mathcal{H}\,\mathrm{e}^{-(\mathrm{i}/\hbar)\mathcal{H}t}C\rho(\beta_{\mathrm{i}})]\\&=\mathrm{Tr}\,[C^{\dagger}\mathcal{H}C\rho(\beta_{\mathrm{i}})]\,.\end{aligned} \tag{A.6}$$

Hence eq. (A.1) takes on the form

$$\mathrm{Tr}[\mathcal{H}\rho(\beta)]=\lim_{t\to-\infty}\mathrm{Tr}[\mathcal{H}\rho(t)]=\mathrm{Tr}[\mathcal{H}\rho(\beta_{\mathrm{i}})]+\int_0^{-\infty}\mathrm{Tr}\Big(\mathcal{H}\,\frac{\mathrm{d}}{\mathrm{d}t'}\,\rho(t')\Big)\,\mathrm{d}t'\,, \tag{A.7}$$

where

$$\rho(t)=U(t)\;\rho(\beta_{\mathrm{i}})\;U(t)^{\dagger}\,. \tag{A.8}$$

The last term in eq. (A.7) may be transformed to give

$$\begin{aligned}\int_0^{-\infty}\mathrm{Tr}\Big(\mathcal{H}\,\frac{\mathrm{d}}{\mathrm{d}t'}\,\rho(t')\Big)\,\mathrm{d}t'&=\int_0^{-\infty}\mathrm{Tr}\Big([\mathcal{H}-\mathcal{H}(t,h)]\,\frac{\mathrm{d}}{\mathrm{d}t'}\,\rho(t')\Big)\,\mathrm{d}t'\\&=\mathrm{Tr}\{[\mathcal{H}(0,h)-\mathcal{H}]\rho(\beta_{\mathrm{i}})\}+\int_0^{-\infty}\mathrm{Tr}\Big(\frac{\partial\mathcal{H}(t',h)}{\partial t'}\,\rho(t')\Big)\,\mathrm{d}t'\,,\end{aligned} \tag{A.9}$$

so that the condition (A.1) for β reads

$$\mathrm{Tr}[\mathcal{H}\rho(\beta)] = \mathrm{Tr}[\mathcal{H}(0,h)\,\rho(\beta_i)] + \int_0^{\infty} \mathrm{Tr}\left(\frac{\partial \mathcal{H}(t',h)}{\partial t'}\,\rho(t')\right) \mathrm{d}t' \,. \tag{A.10}$$

The energy change $\langle \partial\mathcal{H}/\partial t'\rangle(t') = \langle \partial\mathcal{H}_S/\partial t'\rangle(t')$ is proportional to the number of particles of the subsystem N_S. Hence in the thermodynamic limit of the bath we obtain

$$\begin{aligned}\lim_{N_B\to\infty} \frac{1}{N_B} \mathrm{Tr}[\mathcal{H}\rho(\beta)] &= \lim_{N_B\to\infty} \frac{1}{N_B} \mathrm{Tr}\{[\mathcal{H} + \mathcal{H}^1_S(h,0)]\}\,\rho(\beta_i) \\ &= \lim \frac{1}{N_B} \mathrm{Tr}[\mathcal{H}\rho(\beta_i)] \,,\end{aligned} \tag{A.11}$$

or

$$\beta = \beta_i \,.$$

Appendix B

Decay of response functions

The system is again taken to consist of a subsystem coupled to a heat bath. We will deduce the properties of the response functions from special fields.

Let fields act upon the system between times $t = 0$ and $t = t^*$. At time $t = 0$ we start from equilibrium. Then at times $t - t^*$ longer than a relaxation time τ_R [#6] equilibrium is restored. Due to the large heat capacity of the system no change in temperature is to be considered. Thus we have

$$\beta = \beta_i \qquad \text{for } t - t^* \gg \tau_R \,. \tag{B.1}$$

Especially if we take fields consisting of n δ-pulses,

$$L^1(t) = \left(\sum_m c_m L_m\right) \sum_{\alpha=1}^{n} h_\alpha \delta(t - \tau_\alpha) \,, \tag{B.2}$$

with

$$\tau_n > \tau_{n-1} > \cdots > \tau_1 > 0 \tag{B.3}$$

[#6] Here τ_R just denotes that relaxation times exist. We do not mention their dependence on observables and the state $\rho(0)$.

(c_m arbitrary factors), then for observation times $t - \tau_n \gg \tau_R$ an expectation value of an observable F of the subsystem must be given by its equilibrium value

$$\langle F\rangle(t) = \langle F\rangle_{\beta_i}, \qquad \text{for } t - \tau_n \gg \tau_R . \tag{B.4}$$

Further, if the time difference between any two neighbouring pulses is longer than τ_R,

$$\tau_v - \tau_{v-1} \gg \tau_R , \tag{B.5}$$

equilibrium is also reached at a time $t = \tau_v - 0$,

$$\rho(\tau_v - 0) = \rho_{\beta_i} . \tag{B.6}$$

This means that in case (B.5) the expectation value $\langle F\rangle(t)$ for $t > \tau_v$ cannot depend on the factors $h_1, h_2, \ldots, h_{v-1}$. Thus we have

$$\langle F\rangle(t) = f(t, h_v, \ldots, h_n), \qquad t > \tau_n . \tag{B.7}$$

Transforming the properties (B.4) and (B.7) into the language of response we will obtain the desired results.

Take the special interaction (B.2) then the contribution to the nth order response with the factor $h_1 h_2 \cdots h_n$ is found from eq. (4.7) to be

$$\langle F\rangle(t)^{(n)} = \cdots + h_1 h_2 \cdots h_n \varphi(t - \tau_n, \tau_n - \tau_{n-1}, \ldots \tau_2 - \tau_1), \qquad t > \tau_n , \tag{B.8}$$

where the functions φ are defined by

$$\varphi(t_n, \ldots, t_1) = \sum_{m_1 \cdots m_n} c_{m_1} \ldots c_{m_n} \operatorname{Tr}(F\, e^{-iLt_n} L_{m_n} e^{-iLt_{n-1}} L_{m_{n-1}} \cdots e^{-iLt_1} L_{m_1} \rho_{\beta_i}) . \tag{B.9}$$

From eq. (B.4) we see that this contribution must vanish for $t - \tau_n \gg \tau_R$. The same is true for a particular v satisfying eq. (B.5) since in $\langle F\rangle(t)^{(n)}$ no functions of $h_1, \ldots, h_{v-1}$ may occur [eq. (B.7)]. Hence we get the result that

$$\varphi(t_n, \ldots, t_1) \to 0, \qquad t_v \gg \tau_R , \tag{B.10}$$

holds for any particular t_v larger than τ_R. The factors c_m are arbitrary, so that the traces in eq. (B.9) have the same properties as eq. (B.10).

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