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1971 J. Phys. A: Gen. Phys. 4 555

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A general approach to nonequilibrium quantum statistics

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Abstract. Direct use of a generalized Laplace transform is proposed for applications in nonequilibrium quantum statistics. The method includes the equilibrium 'double-time' Green's function method as a special case.

An example is given in optical theory in which non-Markoffian behaviour and Poincaré recurrences have been explicitly found.

1. Introduction

The method of thermodynamic 'double-time' Green's functions has been very successful in dealing with problems in equilibrium statistical physics and linear-response theory (Zubarev 1960). Once the Green's function G(A; B) of two operators A and B is known, correlation functions such as

$$F(A; B) = \langle A(t)B(0) \rangle_{eq}$$
(1)

can be found. Here

$$\langle x \rangle_{eq} = \operatorname{Tr}(\rho_{eq} x)$$

 $A(t) = \exp(\mathrm{i}Ht) A \exp(-\mathrm{i}Ht)$

the operator A in the Heisenberg representation, $\rho_{eq} = \exp(-\beta H)/Z$, the canonical density matrix, Z is the partition function

and

$$Z = \operatorname{Tr} \{ \exp(-\beta H) \}$$
$$\beta = \frac{1}{kT}$$

where k is Boltzmann's constant and T is the absolute temperature. H, the Hamiltonian of the system, is assumed to have no explicit time dependence. In this paper we outline a method of tackling general non-equilibrium problems in statistical physics which is closely related to the equilibrium Green's function method. In fact G(A; B) can be obtained from this approach and thus one can recover the usual results for equilibrium expectation values and linear susceptibilities. In the nonequilibrium situation the method can be used to calculate diagonal elements of the density matrix via the generalized master equation, but in general provides a more powerful technique than the master equation for direct calculation of quantum statistical averages. We demonstrate this in an example which shows non-Markoffian behaviour and Poincaré recurrences.

The method uses generalized Laplace or Fourier transforms (Titchmarsh 1937) and for later reference we present the main formulae here. We define the generalized Laplace transform of a function f(t) to be

$$L(\omega) = -i \int_{0}^{\infty} dt f(t) \exp(i\omega t)$$

$$\omega = u + iv \qquad v \ge 0$$
555
(2)

if a sufficiently large v can be found for the integral to exist. The reciprocal formula is

$$f(t) = -\frac{1}{2\pi i} \int_{-\infty+ia}^{\infty+ia} d\omega L(\omega) \exp(-i\omega t) \qquad a, t > 0$$
(3)

for sufficiently large a. The advantage of Laplace transform techniques in quantum mechanics are much the same as in, for instance, electrical engineering, namely: (i) they convert systems of differential equations into systems of algebraic equations; and (ii) they build boundary conditions into the algebraic equations in a convenient manner.

In the next section we outline briefly the thermodynamic double-time Green's function equation of motion technique. In § 3 we show how more general problems in statistical physics can be approached by a closely related but simpler technique which makes direct use of the generalized Laplace transform. We also give a simple illustrative example. In the fourth section we apply the technique to a problem in optics which has proved difficult and lengthy to solve by other techniques. In the final section we draw some general conclusions.

2. The Green's function technique

The retarded Green's function, $G^+_{\omega}(A; B)$, of two operators A and B in the complex-frequency domain is obtained by applying the transform (2) to the function

$$G_i(A; B) = \langle [A(t), B(0)]_\eta \rangle_{eq}$$
(4)

where $[X, Y]_{\eta} = XY - \eta YX$, and η can have the values ± 1 . When η is ± 1 we shall speak of Bose Green's functions, and when $\eta = -1$ of Fermi Green's functions. It will be apparent later why one chooses to work with the Laplace transform of the η commutator, rather than that of the correlation function directly. Introducing the exact eigenstates $|n\rangle$, the corresponding exact eigenvalues, E_n of the Hamlitonian H and writing out the trace in (4) explicitly using ρ_{eq} we have

$$G_t(A;B) = Z^{-1} \sum_{n,m} \{ \exp(-\beta E_n) - \eta \exp(-\beta E_m) \} A_{nm} B_{mn} \exp(iE_{nm}t)$$
(5)

and

$$G_{\omega}^{+}(A; B) = -i \int_{0}^{\infty} dt \exp(i\omega t) G_{t}(A; B)$$

= $Z^{-1} \sum_{n,m} \frac{\exp(-\beta E_{n}) - \eta \exp(-\beta E_{m})}{(\omega + E_{nm})} A_{nm} B_{mn}$ (6)
where $E_{nm} = E_{n} - E_{m}.$

where

Using the explicit form of $G^+_{\omega}(A; B)$, it is very easy to show that it satisfies the equation

$$\omega G^+_{\omega}(A;B) = \langle [A,B]_{\eta} \rangle_{\text{eq}} + G^+_{\omega}([A,H];B)$$
(7)

which we shall refer to as the equation of motion for $G^+_{\omega}(A; B)$. When we do not depict the time explicitly in an operator in the Heisenberg representation, we mean that operator at time zero. Thus $\langle [A;B]_n \rangle_{eq} \equiv \langle [A(0), B(0)]_n \rangle_{eq}$. $G^+_{\omega}(A;B)$ is usually calculated by writing down its equation of motion, then the equation for $G^+_{\omega}([A, H]; B)$, etc., so generating a hierarchy of equations which can be truncated by some approximation scheme and so solved for $G^+_{\omega}(A; B)$. We now see the reason for introducing the η commutator into (4)—it results in our having to calculate the average $\langle [A, B]_n \rangle_{eq}$ in (7). Now the η commutator of two operators is frequently much simpler than the two operators taken alone (in many cases it is one or zero), so that the average $\langle [A, B]_n \rangle_{eq}$ can be evaluated much more easily than the average $\langle AB \rangle_{eq}$. However, this device causes a complication when one comes to consider the inverse transform (3), as a straightforward application of it would lead to (4), and not to $\langle A(t)B(0) \rangle$ which is normally required. The correct inversion formula for the η -commutator Green's function is

$$\langle A(t)B(0)\rangle_{eq} + (1+\eta)C(AB) = -\frac{1}{2\pi i} \oint d\omega \frac{G_{\omega}^{r}(A;B)\exp(-i\omega t)}{1-\eta\exp(-\beta\omega)} \qquad t > 0$$
(8)

where the contour encloses the real axis.

The constant contribution C(AB), which only appears for Bose Green's functions, is the origin of the so-called zero-frequency anomaly (see, for example, Stevens and Toombs 1965, Lucas and Horwitz 1969, Kwok and Schultz 1969). It arises from the zero of $\{1 - \eta \exp(-\beta\omega)\}$ at $\omega = 0$ when $\eta = +1$.

By substituting from (6) into (8), one sees the need for the $\{1 - \eta \exp(-\beta\omega)\}$ factor. The inversion (8) is based implicitly on the Gibb's weighting provided by the use of the canonical density matrix.

3. The generalized Laplace-transform technique

When one comes to consider non-equilibrium problems, that is, problems described statistically by an arbitrary density matrix rather than by the canonical density matrix $\rho_{eq} = \exp(-\beta H)/Z$, the situation is different. There are two main types of problem:

(i) When the initial expectation value of some operator is known, and one wishes to calculate how it evolves when a perturbation is applied to the system.

(ii) When one wishes to calculate the steady state (but not equilibrium) value of a correlation function $\langle A(t)B(0) \rangle$ (this is the situation which arises for example in considering transport or scattering problems). In these situations there is no advantage in using the η commutator and we propose to work with the simple products A(t)B(0). Hence we define

$$L(A, B) = -i \int_{0}^{\infty} dt \langle A(t)B(0) \rangle \exp(i\omega t)$$
(9)

with arbitrary initial ρ , to use instead of the Green's functions of the previous section. Explicitly we find the Laplace transform

$$L(A; B) = \sum_{n,m} \frac{A_{nm}(B\rho)_{mn}}{\omega + E_{nm}}.$$
(10)

It satisfies the equation of motion

$$\omega L(A; B) = \langle AB \rangle + L([A, H]; B).$$
(11)

(Compare with (7).) The inversion formula (3) is used to recover $\langle A(t) B(0) \rangle$ once L(A; B) is known, and of course L(A; B) can be calculated in a similar way to that in which $G_{\omega}(A; B)$ is calculated. We note that in this case the inversion formula involves no term corresponding to the zero-frequency anomaly. To employ this technique we have to know the $\langle AB \rangle$ which are the essential parameters of the

problem. Replacing $\langle AB \rangle$ with the simpler $\langle [A, B]_n \rangle$ as is contrived in the double time Green's function technique does not work in the non-equilibrium case because the former technique requires the density matrix to be explicitly of the Gibb's form.

In the first type of problem where one wishes to follow the time evolution of a single operator, A, say, one can define a function L(A) by applying (2) to the quantity $\langle A(t) \rangle$. Then L(A) obeys

$$\omega L(A) = \langle A \rangle + L([A, H])$$

and formulae (10) and (11) hold also for L(A) if B is replaced by unity.

The basic formulae of the thermodynamic Green's function and generalized Laplace transform methods are compared in table 1. It should be noted that, in the case of thermodynamic equilibrium, the inversion formulae on the last line of the table can be made identical, before integration, by simple manipulation using the Gibbs form of the density matrix. The generalized Laplace transform method, therefore, includes the double-time Green's function method as a special case.

To demonstrate the technique, we consider as a simple illustrative example a system of bosons interacting with a two-level atom through the Hamiltonian

$$H = \sigma_+ \sigma_- \omega_0 + a^+ a \omega_q + g(\sigma_+ a + a^+ \sigma_-).$$
⁽¹²⁾

 a^+ and a obey the usual commutation rules for Bose operators, and σ_+ and σ_- are the Pauli operators. The final term represents the interaction between the atom and the field in the so-called rotating wave approximation. For a discussion of this Hamiltonian see, for example, Sears (1964). It is relevant to a single phonon interacting with a paramagnetic impurity, or a photon interacting with an atom. We suppose that there is some external agency (a reservoir) which maintains the operator $\sigma_3 = (\sigma_+ \sigma_- - \frac{1}{2})$ constant on a time scale in which changes in a(t) can occur. Our Hamiltonian then describes a boson field interacting with a type of heat bath, as energy can be transferred to and from the field without affecting the energy of the atom. We calculate the mean boson occupation number $\langle a^+(t)a(t) \rangle$ as a function of time with a view to investigating whether a steady state solution can exist. If the steady state can exist, and if we choose our initial conditions suitably, we will find that $\langle a^+(t)a(t) \rangle$ will be independent of time. We calculate the quantity $L(a^+a)$ using (11). We find

$$\omega L(a^+a) = \langle a^+a \rangle + gL(\sigma_-a^+ - a\sigma_+)$$
(13)

$$(\omega + \omega_q - \omega_0) L(\sigma_a^+) = -gL(\sigma_+ \sigma_- + 2\sigma_3 a^+ a) \sim -g\left(\frac{\langle \sigma_+ \sigma_- \rangle}{\omega + 2\langle \sigma_3 \rangle L(a^+ a)}\right)$$
(14)

$$(\omega - \omega_q + \omega_0) L(\sigma_+ a) = gL(\sigma_+ \sigma_- + 2\sigma_3 a^+ a) \sim g\left(\frac{\langle \sigma_+ \sigma_- \rangle}{\omega + 2\langle \sigma_3 \rangle L(a^+ a)}\right)$$
(15)

when we use the fact that σ_3 is constant and that $L(1) = 1/\omega$. We have also assumed $\langle \sigma_- a^+ \rangle = \langle \sigma_+ a \rangle = 0$ for simplicity. With this approximation we can solve the system of equations to find

$$L(a^{+}a) = \frac{\langle a^{+}a \rangle (\omega^{2} - \omega_{q0}^{2}) - 2g^{2} \langle \sigma_{+}\sigma_{-} \rangle}{\omega(\omega^{2} - \omega_{q0}^{2} + 4g^{2} \langle \sigma_{3} \rangle)}.$$
 (16)

We obtain $\langle a^+(t)a(t) \rangle$ from the inversion formula (3) by closing the contour in the



'l'hermodynamic 'double-time' Green's functions

Generalized Laplace transform

 $\langle A(t)B(0)\,
angle = -rac{1}{2\pi\mathrm{i}}\int_{\mathrm{i}a+\infty}^{\mathrm{i}a+\infty}\mathrm{d}\omega\,L(A;B)$ $L(A, B) = -\mathrm{i} \int_0^\infty \mathrm{dt} \, F(A; B) \exp(\mathrm{i}\omega t)$ $= \sum_{n,m} A_{nm}(B\rho)_{nm} \exp(\mathrm{i}\omega_{nm}t)$ $\omega L(A, B) = \langle AB
angle + L([A, H]; B)$ $= \sum_{n,m} \frac{A_{nm}(B\rho)_{mn}}{\omega + E_{nm}}$ $F(A,\,B)=\left< A(t)B(0)
ight>$ $\left< A(t)B(0) \right>_{\mathrm{eq}} = rac{1}{2\pi\mathrm{i}} \int_{\mathrm{i}a-\infty}^{\mathrm{i}a+\infty} rac{\mathrm{d}\omega G_\omega(A;B) \exp(-\mathrm{i}\omega t)}{1-\eta \exp(-eta\omega)}$ $egin{aligned} G_{t}(A;B) &= \langle [A(t),B(0)]_{n}
angle_{ ext{eq}} \ &= Z^{-1} \sum_{n,m} \left\{ \exp(-eta E_{n}) - \eta \exp(-eta E_{m})
ight\} \end{aligned}$ $=\frac{1}{Z}\sum_{n,m}\frac{\exp(-\beta E_n)-\eta\exp(-\beta E_m)}{\omega+E_{nm}}$ $\omega G_o(A;B) = \langle [A,B]
angle + G_o([A,H];B)$ $G_{\omega}(A, B) = -\mathrm{i} \int_{0}^{\infty} \mathrm{d}t \, G_{i}(AB) \exp(\mathrm{i}\omega t)$ $\times A_{nm}B_{mn}\exp(\mathrm{i}E_{nm}t)$ Equation of motion Frequency domain Inversion formula Time domain

t > 0

 $imes \exp(-\mathrm{i}\omega t)$

t > 0

 $+(1+\eta) C(A,B)$

lower half plane. Hence

$$\langle a^{+}(t)a(t)\rangle = \frac{\{\langle a^{+}a\rangle\omega_{q0}^{2} + 2g^{2}\langle\sigma_{+}\sigma_{-}\rangle - 2g^{2}(2\langle\sigma_{3}\rangle\langle a^{+}a\rangle + \langle\sigma_{+}\sigma_{-}\rangle)\cos\Omega t\}}{\omega_{q0}^{2} - 4g^{2}\langle\sigma_{3}\rangle}$$
(17)

where $\Omega = \{\omega_{q0}^2 - 4g^2 \langle \sigma_3 \rangle\}^{1/2}$ and $4g^2 \langle \sigma_3 \rangle < \omega_{q0}^2$.

To investigate whether a steady state exists we put $\langle a^+(t)a(t) \rangle = \langle a^+a \rangle$ when we obtain

$$\langle a^+a \rangle \left(1 - \frac{\omega_{q0}^2 - 4g^2 \langle \sigma_3 \rangle \cos \Omega t}{\omega_{q0}^2 - 4g^2 \langle \sigma_3 \rangle} \right) = \frac{2g^2 \langle \sigma_+ \sigma_- \rangle (1 - \cos \Omega t)}{\omega_{q0}^2 - 4g^2 \langle \sigma_3 \rangle}$$

or

$$\langle a^+a\rangle = -\frac{\langle \sigma_+\sigma_-\rangle}{2\langle \sigma_3\rangle}.$$
 (18)

Hence a steady state does exist, providing that

 $\langle \sigma_3 \rangle < 0.$

The mean boson occupation number is given by (18). Further, suppose that the atom can be defined by a temperature T such that

$$\langle \sigma_3 \rangle = -\frac{1}{2} \operatorname{tanh}(\frac{1}{2}\beta\omega_0) \qquad \beta = 1/kT.$$

(This follows by assuming a Gibbs distribution for the atomic energy levels). We then find

$$\langle a^+a \rangle = -\frac{\langle \sigma_3 \rangle + \frac{1}{2}}{2 \langle \sigma_3 \rangle} = \frac{1}{\exp(\beta\omega_0) - 1}$$
(20)

which is just the Bose factor. Thus, under these conditions the steady state is the same as the equilibrium state and the atom and field are defined by the same temperature. If the inequality (19) does not hold we have the condition for laser action and to obtain a steady state additional damping and non-linear terms must be taken into account.

4. A two-level atom interacting with all the modes of the radiation field

Here we consider a nontrivial example which has been treated recently in detail by Davidson and Kozak (1969, 1970) using the Prigogine-Resibois master equation. We shall show how to obtain their results much more simply using the generalized Laplace transform technique. The model they considered is a two-level atom in a one-dimensional cavity of length L interacting with all the modes of the radiation field. Initially the atom was assumed to be in its excited state, and the probability of it still being in its excited state at a later time t was calculated. The Hamiltonian is a generalization of the one considered in the previous section:

$$H = \sum_{q} a_{q}^{\dagger} a_{q} \omega_{q} + \sigma_{+} \sigma_{-} \omega_{0} + \sum_{q} g_{q} (a_{q}^{\dagger} \sigma_{-} + a_{q} \sigma_{+})$$
(21)

where $g_q^2 = \alpha c \omega_q / L$, α is a dimensionless coupling constant, c the velocity of light. We wish to calculate $\langle \sigma_+(t)\sigma_-(t) \rangle$, which is the probability of the atom being in its excited state at time t. It is easy to write down the hierarchy of equations for $L_{\omega}(\sigma_+\sigma_-)$ using equation (11) and the known commutation relations of the Bose and Pauli operators:

$$\omega L(\sigma_+\sigma_-) = \langle \sigma_+\sigma_- \rangle + \sum_q g_q L(-a_q^+\sigma_- + a_q\sigma_+).$$
(22)

$$(\omega + \omega_{q0})L(a_q^+ \sigma^-) = -g_q L(\sigma_+ \sigma_-) - 2\sum_{q_1} g_q L(\sigma_3 a_q^+ a_{q_1})$$
(23)

$$(\omega - \omega_{q0})L(a_{q}\sigma^{+}) = g_{q}L(\sigma_{+}\sigma_{-}) + 2\sum_{q_{1}} g_{q_{1}}L(\sigma_{3}a_{q_{1}}^{+}a_{q})$$
(24)

$$(\omega + \omega_{qq_1})L(\sigma_3 a_q^+ a_{q_1}) = \frac{1}{2} \{ g_q L(a_{q_1} \sigma_+) - g_{q_1} L(a_q^+ \sigma_-) \} + \sum_{q_2} g_{q_2} L(a_q^+ a_{q_2}(a_{q_1} \sigma_+) - (\sigma_- a_q^+) a_{q_2}^+ a_{q_1}).$$
(25)

To decouple these equations, we assume, in (22) and (24) that

$$L(\sigma_3 a_q^+ a_{q_1}) \simeq L(\sigma_3 a_q^+ a_q) \delta_{qq_1}$$

and in (25), that

$$L(a_{q}^{+}a_{q_{2}}(a_{q_{1}}\sigma_{+})) \simeq L(a_{q}^{+}a_{q}(a_{q_{1}}\sigma_{+}))\delta_{qq_{2}} + L(a_{q}^{+}a_{q}(a_{q_{2}}\sigma_{+}))\delta_{qq_{1}} - L(a_{q}^{+}a_{q}a_{q}\sigma_{+})\delta_{qq_{1}}\delta_{qq_{2}}$$
(26)

This is a reasonable approximation at least in the weak coupling region when one can still think of bosons and atoms as separate identities. Now $L(a_q^+a_q(a_q\sigma_+))$ is related to $\langle n_q(t)a_q(t)\sigma^+(t)\rangle$ where $n_q(t) = a_q^+(t)a_q(t)$ so that if n_q is always small,

$$L(a_q^+a_q(a_q\sigma_+)) \ll L(a_q\sigma_+).$$
⁽²⁷⁾

With the stated initial conditions $\langle n_q \rangle$ satisfies the identify $\Sigma_q \langle n_q(t) \rangle \leq 1$, so that if the number of modes is large the occupation of any particular mode will be small, i.e. $\langle n_q(t) \rangle \ll 1$, and (27) holds. A similar argument holds for the other terms in (26), and so the final term in (25) can be neglected compared with the first term. Hence (23), (24) and (25) become

$$(\omega + \omega_{q0})L(a_q^+ \sigma_-) = -g_q L(\sigma_+ \sigma_-) - 2g_q L(\sigma_3 a_q^+ a_q)$$
(28)

$$(\omega - \omega_{q0})L(a_q\sigma_+) \simeq g_q L(\sigma_+\sigma_-) + 2g_q L(\sigma_3 a_q^+ a_q)$$
⁽²⁹⁾

$$\omega L(\sigma_3 a_q^+ a_q) \simeq \frac{1}{2} g_q L(a_q \sigma_+ - a_q^+ \sigma_-).$$
(30)

Equations (22), (28), (29) and (30) are then easily solved to give

$$L(\sigma_{+}\sigma_{-}) = \frac{\langle \sigma_{+}\sigma_{-} \rangle}{\omega [1 - 2 \Sigma_{q} g_{q}^{2} / (\omega^{2} - \omega_{q0}^{2} - 2g_{q}^{2})]}.$$
 (31)

Now for an infinite system, $g_q^2 \to 0$ (although $\Sigma_q g_q^2 \neq 0$) and with the initial condition $\langle \sigma_+ \sigma_- \rangle = 1$, (31) reduces in this limit to

$$L(\sigma_{+}\sigma_{-}) = \left\{\omega\left(1 - 2\sum_{q} \frac{g_{q}^{2}}{\omega^{2} - \omega_{q0}^{2}}\right)\right\}^{-1}$$
(32)

which is the main result of Davidson and Kozak's paper. The sum on the right hand

561

side is related to one which occurs in the theory of the Lamb shift. For a discussion of the inversion of (32), and the physical properties of $\langle \sigma_+(t)\sigma_-(t) \rangle$, including Poincaré recurrences and non-Markoffian behaviour, we refer to Davidson and Kozak (1969, 1970). We merely wish to point out here the comparative simplicity of our derivation. Furthermore, our result (31), as opposed to (32), also gives the exact results for a single mode when L is finite (see for example, Fleck 1967 for a derivation of the exact result in this case). It is clear that the generalized Laplace transform technique can also be extended in a straightforward manner to give results to higher accuracy. It is not so easy to do this in the master-equation approach.

5. Conclusions

We note the following points in conclusion:

(i) The quantity η does not appear in the definition of the *L*-functions so that the same definitions suffices for both fermions and bosons.

(ii) Having solved for L(A; B) by the equation of motion method one finds it to be a function of several of the initial average values such as $\langle AB \rangle$. Normally only a few such averages are required, and this demonstrates the economy of the method; only those initial conditions which are relevant to the problem in hand and the level of approximation required are introduced. One does not need an explicit knowledge of the density matrix. Contrast this with the master equation approach where one has to use the initial elements of the density matrix to determine the density matrix at time t, and then perform a trace with the operator whose mean value is wanted. One is reminded of the approach of Fano (1957) who regards the density matrix as being defined by the average values of a sufficient number of suitably chosen operators, rather than directly by the density matrix elements themselves. In this sense one retains only the relevant density matrix elements in the generalized Laplace transform method.

(iii) By comparing the equations of motion for G_{ω} and L (equations (7) and (11)) one sees they differ only in that $\langle [A, B] \rangle_{eq}$ in (7) is replaced by $\langle AB \rangle$ in (11). Thus one can obtain G_{ω} from L by replacing average values whenever they occur by the corresponding η -commutator average. (It is not so easy to obtain L from G_{ω} simply because the commutators $\langle [A, B]_{\eta} \rangle_{eq}$ are frequently one or zero).

(iv) There is no zero-frequency anomaly in the generalized Laplace transform approach because all the singularities in the inversion formula integrand arise from the L-function itself unlike the case of the Bose Green's function.

(v) In the steady-state case, the quasi-particle energies and damping can be obtained from the poles of a suitable L-function, just as is frequently done in the equilbrium case from the poles of the relevant Green's function.

(vi) If one wishes to calculate the density matrix elements, one could do this by calculating L for the projection operator $|s_t\rangle\langle r_t|$. For

$$\langle (|s_t \rangle \langle r_t|) \rangle = \operatorname{Tr}(\rho |s_t \rangle \langle r_t|) = \rho_{rs}(t).$$

A similar approach was taken by Pike (1965) who derived a generalized master equation using projection operators and equation of motion techniques closely related to the ones described here.

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