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Derivation of Generalized Master Equations*†

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We present a simplified derivation of a generalized master equation for the diagonal part of the occupation probability density. This derivation is valid for systems of arbitrary volume. It does not require the use of perturbation expansions nor the use of a diagonal singularity condition. In addition, a similar derivation is presented of a generalized master equation for the nondiagonal part of the occupation probability density. These equations become identical to the generalized master equations of Van Hove and Janner, respectively, if a perturbation expansion is made, if a diagonal singularity condition is assumed, and if the limit of infinite volume is taken.

RECENT advances in the understanding of the nonequilibrium behavior of many-particle quantum systems have been made by Van Hove,[1] by Prigogine and his co-workers,[2] and by others. They have used dynamical arguments to obtain kinetic equations which describe the irreversible evolution of systems from particular initial states. In general, these kinetic equations are non-Markovian, but in some limiting cases they can be approximated by the Markovian Pauli equations.[3] Central in the work of these authors is a many-body perturbation theory applied to an infinite system. In particular, Van Hove has obtained a generalized master equation for the diagonal part of the occupation probability density (g.m.e.d.) by making a perturbation expansion and assuming a diagonal singularity condition. Also, the limit of an infinite system is essential to the analysis so that certain terms can be ignored.

It is clearly desirable to derive a g.m.e.d. without recourse to perturbation theory.[4] We wish to present a simplified derivation of a g.m.e.d. by making use of reasoning similar to that of Heitler and others in the treatment of damping theory.[5] This derivation does not make use of perturbation expansions or of Van Hove's diagonal singularity condition and it is valid for systems of finite volume. The difference between our result and Van Hove's will be discussed below.

Recently, Janner[6] has used the reasoning and assumptions of Van Hove to derive a generalized master equation for the nondiagonal part of the occupation probability density (g.m.e.n.). A simplified derivation of a g.m.e.n. is given below and this g.m.e.n. is compared to Janner's.

We will first present a derivation of the following g.m.e.d.:

$$\frac{dP_E(t/\alpha\alpha_0)}{dt} = f_E(t/\alpha)\ \delta(\alpha - \alpha_0) + 2\pi \sum_{\alpha'} \int_0^t dt'[w_E(t - t'/\alpha\alpha')P_E(t'/\alpha'\alpha_0) - w_E(t - t'/\alpha'\alpha)P_E(t'/\alpha\alpha_0)], \tag{1}$$

where $\delta(\alpha - \alpha_0)$ is a Kronecker delta, and the partial transition probability $P_E(t/\alpha\alpha_0)$ is related to the transition probability $P(t/\alpha\alpha_0)$ for $t > 0$ by

$$\int_{-\infty}^{\infty} dEP_E(t/\alpha\alpha_0) = P(t/\alpha\alpha_0) \equiv \langle\alpha_0]\ \exp\ (iHt/\hbar)\ [\alpha\rangle\ \langle\alpha]\ \exp\ (-iHt/\hbar)\ [\alpha_0\rangle, \tag{2}$$

with $P(0/\alpha\alpha_0) = \delta(\alpha - \alpha_0)$ and the quantities w_E and f_E are defined below. (Our notation will be the same as that used by Van Hove and Janner.) The Hamiltonian, which is assumed not to depend explicitly on the time, is written as a sum of an unperturbed part H_0 and a perturbation H_1,

$$H = H_0 + H_1, \tag{3}$$

with

$$H_0\ [\alpha\rangle = \epsilon_\alpha\ [\alpha\rangle, \tag{4}$$

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[1] L. Van Hove, Physica **23**, 441 (1957).
[2] I. Prigogine, *Non-Equilibrium Statistical Mechanics* (Interscience Publishers, Inc., New York, 1962).
[3] I. Prigogine and P. Resibois, Physica **27**, 629 (1961).
[4] Similar motivations appear in R. W. Zwanzig, in *Lectures in Theoretical Physics*, edited by W. E. Brittin, B. W. Downs, and J. Downs (Interscience Publishers, Inc., New York, 1961), vol. III; E. W. Montroll, in *Fundamental Problems in Statistical Mechanics* (North-Holland Publishing Company, Amsterdam, 1962).
[5] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1953), 3rd ed., p. 164.
[6] A. Janner, Helv. Phys. Acta (to be published).

and we use the eigenstates $|\alpha\rangle$ to compute matrix elements.

Our analysis will be based on the properties of the resolvent operator R_l which has been discussed by Van Hove[1,7] and by Hugenholtz,[8] and is defined by

$$R_l = (H - l)^{-1}, \tag{5}$$

where l is a complex number. The resolvent is related to the time unitary transformation operator according to

$$\exp(-iHt/\hbar) = -(2\pi i)^{-1} \int_C dl\, R_l \exp(-ilt/\hbar), \tag{6}$$

where the contour C is taken counterclockwise around a sufficiently large portion of the real axis.

Introducing Eq. (6) into Eq. (2), we obtain

$$P(t/\alpha\alpha_0) = -(2\pi)^{-2} \int_C dl\, dl' \times \exp[i(l - l')t/\hbar] \langle\alpha_0] R_l [\alpha\rangle \langle\alpha] R_{l'} [\alpha_0\rangle,$$

and defining a quantity X,

$$X_{l'l}(\alpha_0\alpha) = X_{ll'}(\alpha\alpha_0) \equiv \langle\alpha_0] R_l [\alpha\rangle \langle\alpha] R_{l'} [\alpha_0\rangle, \tag{7}$$

we have

$$P(t/\alpha\alpha_0) = -(2\pi)^{-2} \int_C dl\, dl' \times \exp[i(l - l')t/\hbar]\, X_{ll'}(\alpha\alpha_0). \tag{8}$$

The g.m.e.d. is a mathematical consequence of the following equation for X:

$$(l - l')X_{ll'}(\alpha\alpha_0) = F_{ll'}(\alpha)\, \delta(\alpha - \alpha_0) - i \sum_{\alpha'} [\tilde{W}_{ll'}(\alpha\alpha')X_{ll'}(\alpha'\alpha_0) - \tilde{W}_{ll'}(\alpha'\alpha)X_{ll'}(\alpha\alpha_0)], \tag{9}$$

where we have introduced

$$\tilde{W}_{ll'}(\alpha\alpha') \equiv iF_{ll'}(\alpha)W_{ll'}(\alpha\alpha') \tag{10}$$

and F and W are defined below.

We first derive this equation for X and then show how the g.m.e.d. follows from it. In the representation furnished by H_0 we write the resolvent as the sum of its diagonal and nondiagonal parts; e.g. (the complex arguments l and l' will not be explicitly indicated except when needed for clarity)

$$R = R_{\rm d} + R_{\rm nd}, \tag{11}$$

d and nd denoting the diagonal and nondiagonal parts, respectively. Defining D and U by $R_{\rm d} = D$ and $R_{\rm nd} = DUD$ (note that U is nondiagonal), we can write

$$R = (I + DU)\, D, \tag{12}$$

where I is the unit operator. Equation (12) can be rearranged to yield

$$I = (H_0 + H_1 - l)(I + DU)\, D, \tag{13}$$

and if the diagonal part of this equation is taken, we obtain an expression for D,

$$D_l = (H_0 + G_l - l)^{-1}, \tag{14}$$

with

$$G_l = [H_1 + H_1\, D_l U_l]_{\rm d}. \tag{15}$$

If the nondiagonal part of Eq. (13) is taken, we obtain

$$0 = [H_1 + (H_0 - l)\, DU + H_1\, DU]_{\rm nd},$$

and using Eq. (14) we can write

$$U = -[H_1 + H_1\, DU - G\, DU]_{\rm nd}.$$

Substitution of Eq. (15) into this expression yields

$$U = -[H_1 + [H_1]_{\rm nd}\, DU - [H_1\, DU]_{\rm d}\, DU]_{\rm nd}. \tag{16}$$

We find after substitution of Eq. (12) into Eq. (7) and recalling that U is nondiagonal

$$X_{ll'}(\alpha\alpha_0) = D_l(\alpha)\, D_{l'}(\alpha)[\delta(\alpha - \alpha_0) + D_l(\alpha_0)\, D_{l'}(\alpha_0)\, \langle\alpha_0]\, U_l\, [\alpha\rangle \langle\alpha]\, U_{l'}\, [\alpha_0\rangle], \tag{17}$$

with $D_l(\alpha) \equiv \langle\alpha] D_l [\alpha\rangle$. An identity for $D_l(\alpha)\, D_{l'}(\alpha)$ is obtained from Eq. (14):

$$\begin{aligned} F_{ll'}(\alpha) &\equiv D_l(\alpha) - D_{l'}(\alpha) \\ &= [\epsilon_\alpha + G_l(\alpha) - l]^{-1} - [\epsilon_\alpha + G_{l'}(\alpha) - l']^{-1} \\ &= D_l(\alpha)\, D_{l'}(\alpha)[l - l' - G_l(\alpha) + G_{l'}(\alpha)], \end{aligned}$$

which can be solved to give

$$D_l(\alpha)\, D_{l'}(\alpha) = F_{ll'}(\alpha)[l - l' - G_l(\alpha) + G_{l'}(\alpha)]^{-1}. \tag{18}$$

Substitution of Eq. (18) into Eq. (17) yields, after some simplification,

$$(l - l')X_{ll'}(\alpha\alpha_0) = F_{ll'}(\alpha)\, \delta(\alpha - \alpha_0) + F_{ll'}(\alpha)\, D_l(\alpha_0)\, D_{l'}(\alpha_0) U_l(\alpha_0\alpha) U_{l'}(\alpha\alpha_0) + [G_l(\alpha) - G_{l'}(\alpha)]X_{ll'}(\alpha\alpha_0), \tag{19}$$

where we introduced $\langle\alpha_0]\, U_l\, [\alpha\rangle \equiv U_l(\alpha_0\alpha)$, etc. Let us define an operator W by the equation

[7] L. Van Hove, in *La théorie des gas neutres et ionisés*, edited by C. DeWitt and J. G. Detoeuf (John Wiley & Sons, Inc., New York, 1959).

[8] N. M. Hugenholtz, in *Lectures in Theoretical Physics*, edited by W. E. Brittin and B. W. Downs (Interscience Publishers, Inc., New York, 1960), Vol. II.

$$W_{ll'}(\alpha\alpha_0) = U_l(\alpha_0\alpha)U_{l'}(\alpha\alpha_0) - \sum_{\alpha'} W_{ll'}(\alpha\alpha') \times D_l(\alpha')\, D_{l'}(\alpha')U_l(\alpha_0\alpha')U_{l'}(\alpha'\alpha_0). \quad (20)$$

This equation can be iterated to yield

$$W_{ll'}(\alpha\alpha_0) = U_l(\alpha_0\alpha)U_{l'}(\alpha\alpha_0) - \sum_{\alpha'} U_l(\alpha'\alpha)U_{l'}(\alpha\alpha') \times D_l(\alpha')\, D_{l'}(\alpha')U_l(\alpha_0\alpha')U_{l'}(\alpha'\alpha_0) + \cdots, \quad (21)$$

from which it is evident that W has the symmetry

$$W_{ll'}(\alpha\alpha_0) = W_{l'l}(\alpha_0\alpha). \quad (22)$$

We can obtain an expression for the second quantity on the right-hand side of Eq. (19) in terms of X and W as follows: Multiply Eq. (20) by $D_l(\alpha_0)\, D_{l'}(\alpha_0)$ and rearrange terms to obtain

$$\sum_{\alpha'} W_{ll'}(\alpha\alpha')\, D_l(\alpha')\, D_{l'}(\alpha')[\delta(\alpha' - \alpha_0) + D_l(\alpha_0)\, D_{l'}(\alpha_0)U_l(\alpha_0\alpha')U_{l'}(\alpha'\alpha_0)] = D_l(\alpha_0)\, D_{l'}(\alpha_0)U_l(\alpha_0\alpha)U_{l'}(\alpha\alpha_0),$$

which with the aid of Eq. (17) gives the desired relation

$$\sum_{\alpha'} W_{ll'}(\alpha\alpha')X_{ll'}(\alpha'\alpha_0) = D_l(\alpha_0)\, D_{l'}(\alpha_0)U_l(\alpha_0\alpha)U_{l'}(\alpha\alpha_0). \quad (23)$$

This enables us to write Eq. (19) as

$$(l - l')X_{ll'}(\alpha\alpha_0) = F_{ll'}(\alpha)\, \delta(\alpha - \alpha_0) + F_{ll'}(\alpha) \sum_{\alpha'} W_{ll'}(\alpha\alpha')X_{ll'}(\alpha'\alpha_0) + [G_l(\alpha) - G_{l'}(\alpha)]X_{ll'}(\alpha\alpha_0). \quad (24)$$

Now we obtain an expression for $[G_l(\alpha) - G_{l'}(\alpha)]$ in terms of $W_{ll'}$. From the definition of R_l, it follows that

$$R_l - R_{l'} = (H - l)^{-1} - (H - l')^{-1} = R_{l'}R_l(l - l'),$$

and by taking a diagonal element we obtain

$$F_{ll'}(\alpha) = (l - l') \sum_{\alpha_0} X_{ll'}(\alpha\alpha_0). \quad (25)$$

If we sum Eq. (24) over α_0 and use Eq. (25), we obtain

$$\sum_{\alpha'} F_{ll'}(\alpha')W_{ll'}(\alpha\alpha') = -[G_l(\alpha) - G_{l'}(\alpha)]. \quad (26)$$

Instead of using this to eliminate $[G_l(\alpha) - G_{l'}(\alpha)]$ in Eq. (24), we will use

$$\sum_{\alpha'} F_{ll'}(\alpha')W_{ll'}(\alpha'\alpha) = -[G_l(\alpha) - G_{l'}(\alpha)]. \quad (26')$$

Equation (26′) is obtained as follows: We interchange the complex arguments l and l' in Eq. (17) and follow the same analysis that led to Eq. (26). This will yield

$$\sum_{\alpha'} F_{l'l}(\alpha')W_{l'l}(\alpha\alpha') = -[G_{l'}(\alpha) - G_l(\alpha)], \quad (26'')$$

and using the symmetry property of W expressed by Eq. (22) and multiplying by minus one we obtain Eq. (26′).

Using Eq. (26′) to eliminate $[G_l(\alpha) - G_{l'}(\alpha)]$ in Eq. (24), we write

$$(l - l')X_{ll'}(\alpha\alpha_0) = F_{ll'}(\alpha)\, \delta(\alpha - \alpha_0) - i \sum_{\alpha'} [\tilde{W}_{ll'}(\alpha\alpha')X_{ll'}(\alpha'\alpha_0) - \tilde{W}_{ll'}(\alpha'\alpha)X_{ll'}(\alpha\alpha_0)], \quad (27)$$

which is the desired equation, Eq. (9), for X. As indicated by Van Hove,[1] a g.m.e.d. is a mathematical consequence of this equation and for the sake of completeness we demonstrate this in detail in the appendix. In the appendix we also discuss the role played by the inhomogeneous term on the right-hand side of Eq. (27).

Let us now derive a g.m.e.n., which is

$$\frac{dI_E(t/\alpha\alpha'\alpha_0)}{dt} = g_E(t/\alpha\alpha'\alpha_0) + 2\pi \int_0^t dt' \sum_{\alpha''} [w_E(t - t'/\alpha\alpha'')I_E(t'/\alpha''\alpha'\alpha_0) - w_E(t - t'/\alpha''\alpha)I_E(t'/\alpha\alpha'\alpha_0)], \quad (28)$$

$I_E(t/\alpha\alpha'\alpha_0)$ is related to $I(t/\alpha\alpha'\alpha_0)$ for $t > 0$ by

$$\int_{-\infty}^{\infty} dE\, I_E(t/\alpha\alpha'\alpha_0) = I(t/\alpha\alpha'\alpha_0) \equiv \langle\alpha'] \exp(iHt/\hbar)\, [\alpha\rangle\, \langle\alpha] \exp(-iHt/\hbar)\, [\alpha_0\rangle \quad (29)$$

with $I(0/\alpha\alpha'\alpha_0) = 0$, w_E is the same quantity that appears in Eq. (1), and the inhomogeneous term $g_E(t/\alpha\alpha'\alpha_0)$ will be defined below. Introduce Eq. (6) into Eq. (29) to obtain

$$I(t/\alpha\alpha'\alpha_0) = -(2\pi)^{-2} \int_C dl\, dl' \times \exp[i(l - l')t/\hbar]\, Y_{ll'}(\alpha\alpha'\alpha_0), \quad (30)$$

with

$$Y_{ll'}(\alpha\alpha'\alpha_0) \equiv \langle\alpha']\, R_l\, [\alpha\rangle\, \langle\alpha]\, R_{l'}\, [\alpha_0\rangle. \quad (31)$$

The g.m.e.n. (28) is a mathematical consequence of the following equation for Y:

$$(l - l')Y_{ll'}(\alpha\alpha'\alpha_0) = F_{ll'}(\alpha)V_{ll'}(\alpha\alpha'\alpha_0) - i \sum_{\alpha''} [\tilde{W}_{ll'}(\alpha\alpha'')Y_{ll'}(\alpha''\alpha'\alpha_0) - \tilde{W}_{ll'}(\alpha''\alpha)Y_{ll'}(\alpha\alpha'\alpha_0)], \quad (32)$$

where F and $\tilde{W}$ are the same quantities which appear in Eq. (9) and V will be defined below.

We now derive Eq. (32) in essentially the same way we obtained Eq. (27). Substitute Eq. (12) into Eq. (31) to obtain

$$\begin{aligned} Y_{ll'}(\alpha\alpha'\alpha_0) = D_l(\alpha)\, D_{l'}(\alpha)[&D_l(\alpha')U_l(\alpha'\alpha)\;\delta(\alpha-\alpha_0) \\ &+ D_{l'}(\alpha_0)U_{l'}(\alpha\alpha_0)\;\delta(\alpha-\alpha') \\ &+ D_l(\alpha')U_l(\alpha'\alpha)U_{l'}(\alpha\alpha_0)\;D_{l'}(\alpha_0)], \end{aligned} \tag{33}$$

and use Eq. (18) to write

$$\begin{aligned} (l-l')Y_{ll'}(\alpha\alpha'\alpha_0) = F_{ll'}(\alpha)[&D_l(\alpha')U_l(\alpha'\alpha)\;\delta(\alpha-\alpha_0) \\ &+ D_{l'}(\alpha_0)U_{l'}(\alpha\alpha_0)\;\delta(\alpha-\alpha') \\ &+ D_l(\alpha')U_l(\alpha'\alpha)U_{l'}(\alpha\alpha_0)\;D_{l'}(\alpha_0)] \\ &+ [G_l(\alpha)-G_{l'}(\alpha)]Y_{ll'}(\alpha\alpha'\alpha_0). \end{aligned} \tag{34}$$

Let us define an operator V by the equation

$$\begin{aligned} V_{ll'}(\alpha\alpha'\alpha_0) = &D_l(\alpha')U_l(\alpha'\alpha)\;\delta(\alpha-\alpha_0) \\ &+ D_{l'}(\alpha_0)U_{l'}(\alpha\alpha_0)\;\delta(\alpha-\alpha') \\ &+ D_l(\alpha')U_l(\alpha'\alpha)U_{l'}(\alpha\alpha_0)\;D_{l'}(\alpha_0) \\ &- \sum_{\alpha''} D_l(\alpha'')\;D_{l'}(\alpha'') \\ &\times U_l(\alpha''\alpha)U_{l'}(\alpha\alpha'')V_{ll'}(\alpha''\alpha'\alpha_0). \end{aligned} \tag{35}$$

Multiply by $D_l(\alpha)\;D_{l'}(\alpha)$ and rearrange terms to obtain

$$\begin{aligned} \sum_{\alpha''} D_l(\alpha)\;D_{l'}(\alpha)[&\delta(\alpha-\alpha'') + D_l(\alpha'')\;D_{l'}(\alpha'') \\ &\times U_l(\alpha''\alpha)U_{l'}(\alpha\alpha'')]V_{ll'}(\alpha''\alpha'\alpha_0) \\ = D_l(\alpha)\;D_{l'}(\alpha)[&D_l(\alpha')U_l(\alpha'\alpha)\;\delta(\alpha-\alpha_0) \\ &+ D_{l'}(\alpha_0)U_{l'}(\alpha\alpha_0)\;\delta(\alpha-\alpha') \\ &+ D_l(\alpha')U_l(\alpha'\alpha)U_{l'}(\alpha\alpha_0)\;D_{l'}(\alpha_0)]. \end{aligned} \tag{36}$$

The factor multiplying $V_{ll'}(\alpha''\alpha'\alpha_0)$ is recognized from Eq. (17) to be $X_{ll'}(\alpha\alpha'')$ and the right-hand side is recognized from Eq. (33) to be $Y_{ll'}(\alpha\alpha'\alpha_0)$, so we obtain the relation

$$\sum_{\alpha''} X_{ll'}(\alpha\alpha'')V_{ll'}(\alpha''\alpha'\alpha_0) = Y_{ll'}(\alpha\alpha'\alpha_0). \tag{37}$$

Using Eqs. (23) and (37), we write Eq. (35) as

$$\begin{aligned} &D_l(\alpha')U_l(\alpha'\alpha)\;\delta(\alpha-\alpha_0) + D_{l'}(\alpha_0)U_{l'}(\alpha\alpha_0)\;\delta(\alpha-\alpha') \\ &\quad + D_l(\alpha')U_l(\alpha'\alpha)U_{l'}(\alpha\alpha_0)\;D_{l'}(\alpha_0) \\ &= V_{ll'}(\alpha\alpha'\alpha_0) + \sum_{\alpha''} W_{ll'}(\alpha\alpha'')Y_{ll'}(\alpha''\alpha'\alpha_0). \end{aligned} \tag{38}$$

Substituting this into the right-hand side of Eq. (34) and using Eq. (26′) we obtain the desired equation

$$\begin{aligned} (l-l')Y_{ll'}(\alpha\alpha'\alpha_0) = &F_{ll'}(\alpha)V_{ll'}(\alpha\alpha'\alpha_0) \\ &- i\sum_{\alpha''}[\tilde{W}_{ll'}(\alpha\alpha'')Y_{ll'}(\alpha''\alpha'\alpha_0) \\ &- \tilde{W}_{ll'}(\alpha''\alpha)Y_{ll'}(\alpha\alpha'\alpha_0)]. \end{aligned} \tag{39}$$

The g.m.e.n. (28) is a mathematical consequence of this equation, as discussed in the Appendix.

It is thus possible to obtain generalized master equations for the diagonal and nondiagonal parts of the occupation probability density which are valid for finite systems without using perturbation theory and without requiring that the potential satisfy a diagonal singularity condition. Of course in any practical calculation it is convenient and for some things necessary to take the limit of an infinite system and then sums can be replaced by integrals. Furthermore, in this limit the analytic behavior of the resolvent and related operators simplifies, since a set of dense poles along the real axis condense into a branch cut.[1,8]

In order to compare the above results with the work of Van Hove[1,7] and Janner[6] we express the resolvent operator as an expansion in powers of the "interaction" H_1 (see reference 1). If this is done, we obtain the following expression for W:

$$\begin{aligned} W_{ll'}(\alpha\alpha_0) = \{\langle\alpha_0](&H_1 - H_1\,D_lH_1 \\ &+ H_1\,D_lH_1\,D_lH_1 - \cdots)\,[\alpha\rangle\,\langle\alpha] \\ &\times (H_1 - H_1\,D_{l'}H_1 + \cdots)\,[\alpha_0\rangle\}_{id}, \end{aligned} \tag{40}$$

where *id* stands for "irreducible diagonal" and means that all intermediate states are unequal to the initial state and no two intermediate states are equal. Van Hove's expression for W [see Eq. (9.22) in reference 7] is the same as Eq. (40) except for the meaning of *id*. In his work the definition of id is that all intermediate states are "nonidentical" to the initial state and no two intermediate states are "identical"; however, intermediate states may become "occasionally equal." This leads to some terms being counted twice, but Van Hove demonstrates[7] that if a diagonal singularity condition is assumed the error introduced is proportional to one over the volume and thus vanishes in the limit of an infinite system. If we make the same assumptions as Van Hove, then the *id* which appears in Eq. (40) can be replaced by Van Hove's *id*.

We conclude that our results become identical to Van Hove's and Janner's if we adopt their assumptions. However, in some cases it is more convenient to expand the resolvent in terms of some quantity other than the interaction[9]; e.g., if the

[9] R. J. Swenson, J. Math. Phys. (to be published).

interaction contains a hard core it is useful to expand in terms of a scattering matrix.

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APPENDIX

We wish to demonstrate how a g.m.e.d. can be obtained from Eq. (27). The same arguments apply for obtaining Eq. (28) from Eq. (39). From Eq. (8) we have

$$P(t/\alpha\alpha_0) = -(2\pi)^{-2}\int_\gamma dl \int_\gamma dl' \times \exp[i(l-l')t/\hbar]X_{ll'}(\alpha\alpha_0), \tag{A1}$$

and for $t>0$ we can deform the contours to obtain (see reference 7)

$$P(t/\alpha\alpha_0) = (2\pi)^{-2}\int_{-\infty}^{\infty} dE^1 \int_{-\infty}^{\infty} dE^2 \times \exp[i(E^1-E^2-2i\eta)t/\hbar]X_{E^1-i\eta,E^2+i\eta}(\alpha\alpha_0). \tag{A2}$$

Let us make the change of variables

$$E^1 - E^2 = 2E' \quad \text{and} \quad E^1+E^2 = 2E,$$

which yields, for $P(t/\alpha\alpha_0)$,

$$P(t/\alpha\alpha_0) = (2\pi^2)^{-1}\int_{-\infty}^{\infty} dE \int_{-\infty}^{\infty} dE' \times \exp[2i(E'-i\eta)t/\hbar]X_{E+E'-i\eta,E-E'+i\eta}(\alpha\alpha_0), \tag{A3}$$

or

$$P(t/\alpha\alpha_0) = \int_{-\infty}^{\infty} dE\, P_E(t/\alpha\alpha_0), \tag{A4}$$

with $P_E(t)$ defined as

$$P_E(t/\alpha\alpha_0) = (2\pi^2)^{-1}\int_{-\infty}^{\infty} dE' \times \exp[2i(E'-i\eta)t/\hbar]X_{E+E'-i\eta,E-E'+i\eta}(\alpha\alpha_0). \tag{A5}$$

Taking the time derivative of Eq. (A5) yields

$$\frac{dP_E(t/\alpha\alpha_0)}{dt} = i(2\pi^2\hbar)^{-1}\int_{-\infty}^{\infty} dE' \exp[2i(E'-i\eta)t/\hbar] \times 2(E'-i\eta)X_{E+E'-i\eta,E-E'+i\eta}(\alpha\alpha_0), \tag{A6}$$

and from Eq. (27) we obtain an expression for $2(E'-i\eta)\, X_{E+E'-i\eta,E-E'+i\eta}(\alpha\alpha_0)$; i.e.,

$$\begin{aligned}2(E'-i\eta)X_{E+E'-i\eta,E-E'+i\eta}(\alpha\alpha_0) &= F_{E+E'-i\eta,E-E'+i\eta}(\alpha)\,\delta(\alpha-\alpha_0) \\ &\quad - i\sum_{\alpha'}[\tilde{W}_{(\ldots)}(\alpha\alpha')X_{(\ldots)}(\alpha'\alpha_0) \\ &\quad - \tilde{W}_{(\ldots)}(\alpha'\alpha)X_{(\ldots)}(\alpha\alpha_0)],\end{aligned} \tag{A7}$$

where the arguments indicated by $(\cdots)$ are the same as the arguments of F. Equation (A7) enables us to write the time derivative of $P_E(t)$ as

$$\begin{aligned}\frac{dP_E(t/\alpha\alpha_0)}{dt} &= i(2\pi^2\hbar)^{-1}\int_{-\infty}^{\infty} dE' \exp[2i(E'-i\eta)t/\hbar] \\ &\quad\times \{F_{E+E'-i\eta,E-E'+i\eta}(\alpha)\,\delta(\alpha-\alpha_0) \\ &\quad - i\sum_{\alpha'}[\tilde{W}_{(\ldots)}(\alpha\alpha')X_{(\ldots)}(\alpha'\alpha_0) \\ &\quad - \tilde{W}_{(\ldots)}(\alpha'\alpha)X_{(\ldots)}(\alpha\alpha_0)]\}.\end{aligned} \tag{A8}$$

Now we define w_E and f_E by

$$f_E(t/\alpha) = i(2\pi^2\hbar)^{-1}\int_{-\infty}^{\infty} dE' \times \exp[2i(E'-i\eta)t/\hbar]F_{E+E'-i\eta,E-E'+i\eta}(\alpha) \tag{A9}$$

and

$$w_E(t/\alpha\alpha') = (2\pi^2\hbar^2)^{-1}\int_{-\infty}^{\infty} dE' \times \exp[2i(E'-i\eta)t/\hbar]\tilde{W}_{E+E'-i\eta,E-E'+i\eta}(\alpha\alpha') \tag{A10}$$

and we use these definitions in Eq. (A8) to write a g.m.e.d.

$$\begin{aligned}\frac{dP_E(t/\alpha\alpha_0)}{dt} &= f_E(t/\alpha)\,\delta(\alpha-\alpha_0) \\ &\quad + 2\pi\sum_{\alpha'}\int_0^t dt'[w_E(t-t'/\alpha\alpha')P_E(t'/\alpha'\alpha_0) \\ &\quad - w_E(t-t'/\alpha'\alpha)P_E(t'/\alpha\alpha_0)].\end{aligned} \tag{A11}$$

We have made use of the following convolution theorem in obtaining Eq. (A11):

$$\begin{aligned}&\int_0^t dt'\, w_E(t-t'/\alpha\alpha')P_E(t'/\alpha'\alpha_0) \\ &= (4\pi^3\hbar)^{-1}\int_{-\infty}^{\infty} dE' \exp[2i(E'-i\eta)t/\hbar] \\ &\quad\times \tilde{W}_{E+E'-i\eta,E-E'+i\eta}(\alpha\alpha')X_{(\ldots)}(\alpha'\alpha_0).\end{aligned} \tag{A12}$$

Let us prove the above convolution theorem. Substitute the definitions of w_E and P_E into the left-hand side and perform the time integration,

$$\begin{aligned}&\int_0^t dt'\, w_E(t-t'/\alpha\alpha')P_E(t'/\alpha'\alpha_0) \\ &= i(8\pi^4\hbar)^{-1}\mathcal{P}\int_{-\infty}^{\infty} dE'\, dE''\, \tilde{W}_{E+E'-i\eta,E-E'+i\eta}(\alpha\alpha') \\ &\quad\times X_{E+E''-i\eta,E-E''+i\eta}(\alpha'\alpha_0)(E''-E')^{-1} \\ &\quad\times \{\exp[2i(E'-i\eta)t/\hbar] - \exp[2i(E''-i\eta)t/\hbar]\}.\end{aligned}$$

Since the integrand is analytic on the real axis nothing is changed by taking the principal parts $\mathcal{P}$ of the integrals. We do the E'' integration in the first term and the E' integration in the second term to obtain the desired relation (A12). For example, the E'' integration in the first term is

$$B \equiv \mathcal{P} \int dE''\, X_{E+E''-i\eta, E-E''+i\eta}(\alpha'\alpha_0)(E'' - E')^{-1}$$

and closing the contour in the lower half plane (since for large E'', $X \sim E''^{-2}$) we obtain

$$B = \mathcal{P} \int_C dz\, X_{E+z-i\eta, E-z+i\eta}(\alpha'\alpha_0)(z - E')^{-1}.$$

The singularities of $X_{E+z-i\eta, E-z+i\eta}(\alpha'\alpha_0)$ lie above the real E'' axis (as follows from the definition of X), so that the only singularity of the integrand is a simple pole at $z = E'$. The integral is easily evaluated to give (for E' real, we use the definition of the principal part)

$$B = -i\pi X_{E+E'-i\eta, E-E'+i\eta}(\alpha'\alpha_0).$$

The E' integration in the second term is done in exactly the same way and Eq. (A12) is the result.

The derivation of the g.m.e.n. follows in the same manner with g_E defined by

$$g_E(t/\alpha\alpha'\alpha_0) = i(2\pi^2\hbar)^{-1} \int_{-\infty}^{\infty} dE' \times \exp\,[2i(E' - i\eta)t/\hbar] F_{E+E'-i\eta, E-E'+i\eta}(\alpha) \times V_{E+E'-i\eta, E-E'+i\eta}(\alpha\alpha'\alpha_0), \tag{A13}$$

and a convolution theorem identical to Eq. (A12) with I_E and $Y(\alpha\alpha'\alpha_0)$ replacing P_E and $X(\alpha\alpha_0)$.

Van Hove[1] has shown in the limit of weak interaction that the inhomogeneous term in Eq. (A11) serves to specify the initial value of $P(t)$. We wish to show that this is an exact result. To see this, we integrate Eq. (1) over all E and obtain

$$\frac{dP(t/\alpha\alpha_0)}{dt} = \int_{-\infty}^{\infty} dE\, f_E(t/\alpha)\, \delta(\alpha - \alpha_0) + \int_{-\infty}^{\infty} dE \int_0^t dt'[\cdots]. \tag{A14}$$

Designating the inhomogeneous term by Δ and substituting Eq. (A9), we obtain

$$\Delta \equiv \delta(\alpha - \alpha_0) \int_{-\infty}^{\infty} dE\, i(2\pi^2\hbar)^{-1} \int_{-\infty}^{\infty} dE' \times \exp\,[2i(E' - i\eta)t/\hbar] F_{E+E'-i\eta, E-E'+i\eta}(\alpha).$$

From the definition of the resolvent, we obtain the identities

$$R_{E+E'-i\eta} - R_{E-E'+i\eta} = 2(E' - i\eta) R_{E+E'-i\eta} R_{E-E'+i\eta}$$

and

$$\int_{-\infty}^{\infty} dE\, R_{E+E'-i\eta} R_{E-E'+i\eta} = -i\pi(E' - i\eta)^{-1},$$

from which it follows that

$$\int_{-\infty}^{\infty} dE\, [R_{E+E'-i\eta} - R_{E-E'+i\eta}] = -2\pi i.$$

Taking a diagonal part and recalling the definition of $F(\alpha)$,

$$\int_{-\infty}^{\infty} dE\, F_{E+E'-i\eta, E-E'+i\eta}(\alpha) = -2\pi i,$$

allows us to write Δ as

$$\Delta = \delta(\alpha - \alpha_0)(\pi\hbar)^{-1} \int_{-\infty}^{\infty} dE' \exp\,[2i(E' - i\eta)t/\hbar],$$

which reduces to

$$\Delta = \delta(\alpha - \alpha_0)\, \delta(t).$$

Thus, Eq. (A14) becomes

$$\frac{dP(t/\alpha\alpha_0)}{dt} = \delta(\alpha - \alpha_0)\, \delta(t) + \int_{-\infty}^{\infty} dE \int_0^t dt'\, [\cdots], \tag{A15}$$

and it is clear that the inhomogeneous term does not contribute to the time development of $P(t)$.