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Equations of motion in nonequilibrium statistical mechanics of open systems

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In this paper the method of Robertson and that of Zubarev, which have been applied to isolated systems, are modified by using a special projection operator or a special density operator so that they become applicable to open systems. As a result, exact equations are obtained in the form of coupled integro-differential equations with expectation values corresponding to a set of operators of an open system S as the only unknowns. The variables of the system R, which interacts with the system S, are completely eliminated up to the expectation values taken over the initial state of R. The differences between the above mentioned two methods are discussed. It is shown that for special choice of the initial conditions, sets of operators and properties of system R, significant simplifications of the equations of motion can be made. Moreover, an expansion of the equations of motion in powers of the interaction and an approximation of the second order are made. Finally, a Kawasaki-Gunton modification of our projection operator is made in the Appendix.

I. INTRODUCTION

Open systems have been investigated by many authors (see for instance Refs. 1–7). In general, a system S is considered to be open when it interacts with its surroundings, that is to say with any other system R, where the total system $S+R$ is isolated (i.e., the systems S and R are considered to be subsystems of the isolated system $S+R$). In the above-mentioned references the Zwanzig projection technique[8-10] has been used to obtain an equation of motion (EM), the so-called master equation, for the reduced density operator $\rho_S(t)$ of the system of interest S; i.e., the dynamics of the irrelevant system R has been eliminated by an Argyres-Kelley projector, which is time independent.

Some authors[11,12] have indicated the necessity of introducing time-dependent projectors. They have introduced such projectors which no longer eliminate the dynamics of one of the subsystems, but lead to two coupled equations of motion (EM's) for the reduced density operators[11] or for the coarse-grained diagonal parts of the reduced density operators of S and R.[12]

However, in this work we shall limit ourselves only to cases where the dynamics of only one of the subsystems (system S) is of interest and the dynamics of the other subsystem (system R) is to be eliminated. In most cases it is not even necessary to know all the information which is contained in the reduced density operator $\rho_S(t)$ of the system S. Instead, all we need to know are the expectation values (EV's) of a given set of operators (SO's) of the system of interest S: $\{F_n^S,\ n=1,\ldots,m\}$. In none of the above-mentioned articles have the authors obtained closed EM's for such SO's.

We are of the opinion that the problem could most conveniently be solved by modifying the method of Robertson[13,14] or that of Zubarev,[15-18] which have been applied to isolated systems, so that they become applicable to open systems.

Robertson and Mitchell[19] have also briefly treated open systems, namely, they have derived EM's for EV's of two interacting subsystems of an isolated system, where no subsystem has a privileged role, so that the EV's of operators of both subsystems are contained in the equations. It is not possible to decouple these equations in such a way that only the EV's of one subsystem are retained. The decoupling would be of great importance if the dynamics of only one of the sybsystems (open system S) is of interest. Therefore, the equations derived in that paper can not be applied to such cases.

For this reason, in Sec. II we choose a different way by introducing a projection operator $P(t)$, which transforms the time derivative of the density operator $\rho(t)$ of the total system $S+R$ into the time derivative of $\sigma_S(t)\otimes\rho_R(0)$, where $\sigma_S(t)$ is the generalized canonical density operator[13] of the system of interest S, $\rho_R(0)$ is the initial density operator of system R and, as with the most authors, it is assumed that the systems S and R are statistically independent at the initial time $t_0=0$. Moreover, the equation connecting $\rho(t)$ and $\sigma_S(t)\otimes\rho_R(0)$ is derived. Here, as with Robertson,[13,14] an operator $T(t,t')$, which is an integrating factor, is defined. From this connecting equation, we obtain an EM for $\sigma_S(t)\otimes\rho_R(0)$ as well as a system of exact coupled nonlinear integro-differential equations for the EV's of a given SO's of the system of interest S, where the variables of system R are completely eliminated up to the EV's taken over the initial distribution $\rho_R(0)$ of R. This is one of the most important results of our work. We also make an expansion of the integrating factor $T(t,t')$ in powers of the interaction, which is valid, in general, for all time-dependent $P(t)$ operators, which do not necessarily have to have the same form as ours.

Moreover, we come to the important conclusion that it is very advantageous to choose whenever possible sets of S-system operators such that their commutation relations with the unperturbed Hamiltonian of the system S result in linear combinations of the operators of the set. In this case there is a significant simplification of the EM's, especially of the $T(t, t')$ operator in these equations.

In Sec. III, by using a special density operator $\sigma_S(t)\otimes\rho_R(-\infty)$, (the initial time is taken at $t_0=-\infty$), the Zubarev method[15-18] is so modified as to obtain closed EM's for the EV's of the open system S. Some authors[20-22] treating electrical resistivity have applied the Zubarev method to a system interacting with a heat bath. They chose for the irrelevant system R the heat bath and treated the system-bath interaction approximately by introducing a simple relaxation term with a finite inverse relaxation time. Such a relaxation-time-approximation has the disadvantage that it does not always describe the system-bath interaction in a correct way. Contrary to this, we work in this section without introducing any approximations or restrictions regarding the irrelevant system R. Besides, this modified Zubarev method is compared to the modified Robertson method treated in Sec. II and the differences are shown.

In Sec. IV we make some usual assumptions regarding the irrelevant system R. This enables us to make further simplifications in the exact EM's for the EV's $\langle F_n^S\rangle_t$. Moreover, an approximation of second order in the interaction (the so-called Born approximation) is made in these EM's.

In Sec. V we discuss the results obtained. In Appendices A and B we derive some formulas useful for simplifying the EM's. In Appendix C we make a Kawasaki-Gunton modification[23,24] of our operator $P(t)$ in order to obtain a formalism, which more closely resembles the methods using the Zwanzig projection-operator technique.

II. EQUATIONS OF MOTION FOR OPEN SYSTEMS: MODIFIED ROBERTSON METHOD

We consider a system of interest S interacting with a system R, and assume that the total system $S+R$ is isolated, and that its Hilbert space is a direct product $\mathcal{H}_S\otimes\mathcal{H}_R$ of the Hilbert spaces $\mathcal{H}_S$ and $\mathcal{H}_R$ of the systems S and R. Further we suppose that the Hamiltonian of the total system $S+R$ can be written as

$$H=H_0+H_{SR}=\tilde{H}_S+\tilde{H}_R+H_{SR}\,, \tag{2.1}$$

where $H_0=\tilde{H}_S+\tilde{H}_R$, $\tilde{H}_S\equiv H_S\otimes I_R$, $\tilde{H}_R\equiv I_S\otimes H_R$, and H_S, H_R are the Hamiltonians for S and R, acting in $\mathcal{H}_S$ and $\mathcal{H}_R$; I_S, I_R are the unit operators in $\mathcal{H}_S$ and $\mathcal{H}_R$; and H_{SR} is the interaction Hamiltonian, acting in $\mathcal{H}_S\otimes\mathcal{H}_R$. Our Hamiltonian H is assumed to be time independent, but our calculations are also valid for time-dependent Hamiltonians as long as we are not specializing.

We denote the chosen set of S-system operators by $\{F_n^S,\ n=1,\ldots,m\}$, acting in $\mathcal{H}_S$. The spatial dependence of operators F_n^S is not considered, but in this case the calculation can be taken further so far as that an integration over the volume of system S has to be taken besides the summation over n. The operators $\tilde{F}_n^S\equiv F_n^S\otimes I_R$ act in $\mathcal{H}_S\otimes\mathcal{H}_R$. The EV's of the operators $\tilde{F}_n^S$ are given by

$$\langle\tilde{F}_n^S\rangle_t\equiv\mathrm{Tr}_{SR}[\tilde{F}_n^S\rho(t)]\,, \tag{2.2}$$

where Tr_{SR} means that the trace is to be taken over $\mathcal{H}_S$ and $\mathcal{H}_R$, and $\rho(t)$ is the statistical density operator which satisfies the Liouville (von Neumann) equation:

$$i\,\partial\rho(t)/\partial t=(1/\hbar)[H,\rho(t)]=L\rho(t) \tag{2.3}$$

with the normalization $\mathrm{Tr}_{SR}\rho(t)=1$, and L is the Liouville operator.

The reduced density operators corresponding to systems S and R are defined by

$$\rho_S(t)\equiv\mathrm{Tr}_R\rho(t)\,,\quad \rho_R(t)\equiv\mathrm{Tr}_S\rho(t)\,. \tag{2.4}$$

Using $\rho_S(t)$, we may write Eq. (2.2) as

$$\langle\tilde{F}_n^S\rangle_t=\mathrm{Tr}_S[F_n^S\rho_S(t)]\equiv\langle F_n^S\rangle_t\,. \tag{2.5}$$

Now we define the generalized canonical density operator[13,14] for the system of interest S:

$$\sigma_S(t)\equiv\frac{\exp(-\sum_n\lambda_n(t)F_n^S)}{\mathrm{Tr}_S[\exp(-\sum_n\lambda_n(t)F_n^S)]} \tag{2.6}$$

with the normalization $\mathrm{Tr}_S\sigma_S(t)=1$, where the $\lambda_n(t)$ are to be calculated from

$$\langle F_n^S\rangle_t=\mathrm{Tr}_S[F_n^S\sigma_S(t)]\,,\quad n=1,\ldots,m\,. \tag{2.7}$$

We choose the operators $\{I_S, F_n^S;\ n=1,\ldots,m\}$ so that they are linearly independent, i.e.,

$$a_0I_S+\sum_{n=1}^{m}a_nF_n^S=0\,, \tag{2.8}$$

only if $a_0=a_1=\cdots=a_m=0$.

Unlike Robertson, we see no reason for a restriction to the Hermitian operators. When the operators are non-Hermitian one has to take their Hermitian adjoints in the set.

We assume that at the initial time $t=0$, the system S and the system R are statistically independent, so that the density operator factors as

$$\rho(0)=\rho_S(0)\otimes\rho_R(0)\,, \tag{2.9}$$

with $\mathrm{Tr}_S\rho_S(0)=\mathrm{Tr}_R\rho_R(0)=1$. We see from Eqs. (2.5) and (2.7) that either $\rho_S(t)$ or $\sigma_S(t)$ can be used to calculate the EV's $\langle F_n^S\rangle_t$. Since $\sigma_S(t)$ depends on $\lambda_n(t)$ and the latter depends only upon the EV's $\langle F_n^S\rangle_t$, $\sigma_S(t)$ is only a function of the EV's $\langle F_n^S\rangle_t$:

$$\sigma_S(t)=\sigma_S(\langle F_1^S\rangle_t,\ldots,\langle F_m^S\rangle_t)\,, \qquad (2.10)$$

$$\frac{\partial\sigma_S(t)}{\partial t}=\sum_{n=1}^{m}\frac{\partial\sigma_S(t)}{\partial\langle F_n^S\rangle_t}\frac{\partial\langle F_n^S\rangle_t}{\partial t}=\sum_{n=1}^{m}\frac{\partial\sigma_S(t)}{\partial\langle F_n^S\rangle_t}\mathrm{Tr}_{SR}\left(\tilde{F}_n^S\frac{\partial\rho}{\partial t}\right). \qquad (2.11)$$

From this equation it follows that

$$[\partial\sigma_S(t)/\partial t]\otimes\rho_R(0)=(-i)P(t)L\rho(t)\,, \qquad (2.12)$$

where we defined a time-dependent projection operator

$$P(t)A\equiv\sum_n\frac{\partial\sigma_S(t)}{\partial\langle F_n^S\rangle_t}\otimes\rho_R(0)\mathrm{Tr}_{SR}(\tilde{F}_n^SA)\,, \qquad (2.13)$$

with any operator A in the product space $\mathcal{H}_S\otimes\mathcal{H}_R$. It is easy to show that our $P(t)$ operator, although, a modification of Robertson's $P(t)$ operator,[13] has the same properties as Robertson's; i.e., first it holds that

$$\mathrm{Tr}_{SR}[\tilde{F}_n^SP(t)A]=\mathrm{Tr}_{SR}(\tilde{F}_n^SA)\,, \qquad (2.14)$$

where we used the relation which follows from Eq. (2.8),

$$\mathrm{Tr}_S[F_n^S\partial\sigma_S(t)/\partial\langle F_{n'}^S\rangle_t]=\partial\langle F_n^S\rangle_t/\partial\langle F_{n'}^S\rangle_t=\delta_{nn'}\,,$$

where $\delta_{nn'}$ is the Kronecker delta. Second, from Eq. (2.14) it follows that

$$P(t)P(t')A=P(t)A\,. \qquad (2.15)$$

Moreover, in the calculation we must take care that the trace of the operator $P(t)$ is to be taken over all operators which are behind $P(t)$.

By using the Robertson projection-operator formalism[13,14] we obtain from Eq. (2.12) the connecting equation between $\rho(t)$ and $\sigma_S(t)\otimes\rho_R(0)$,

$$\rho(t)-\sigma_S(t)\otimes\rho_R(0)=T(t,0)[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)-i\int_0^t dt'T(t,t')Q(t')L\sigma_S(t')\otimes\rho_R(0)\,, \qquad (2.16)$$

and the EM for $\sigma_S(t)\otimes\rho_R(0)$,

$$[\partial\sigma_S(t)/\partial t]\otimes\rho_R(0)=-iP(t)L\sigma_S(t)\otimes\rho_R(0)-iP(t)LT(t,0)[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)-\int_0^t dt'P(t)LT(t,t')Q(t')L\sigma_S(t')\otimes\rho_R(0)\,, \qquad (2.17)$$

where we defined an integrating operator $T(t,t')$ satisfying the differential equation:

$$\partial T(t,t')/\partial t'=iT(t,t')Q(t')L\,,\quad T(t,t)=I \qquad (2.18)$$

with $Q(t)\equiv I-P(t)$, $I\equiv I_S\otimes I_R$; and $\rho_S(0)$, $\rho_R(0)$ being the given initial density operators.

Introducing the Dyson time-ordering operator $\mathcal{T}$,[25] which orders chronologically from left to right a multiple operator product in sequence of decreasing time, the formal solution of the differential equation for $T(t,t')$, $t\geq t'$ reads

$$T(t,t')=\mathcal{T}\exp\left(-i\int_{t'}^t dt_1Q(t_1)L\right). \qquad (2.19)$$

The expansion in powers of the interaction H_{SR} follows as:

$$T(t,t')=\mathcal{T}\exp\left(-i\int_{t'}^t dt_1T_0(t,t_1)Q(t_1)L_{SR}T_0^{-1}(t,t_1)\right)T_0(t,t')\,, \qquad (2.20)$$

with

$$T_0(t,t')=\mathcal{T}\exp\left(-i\int_{t'}^t dt_1Q(t_1)L_0\right), \qquad (2.21a)$$

and the inverse operator $T_0^{-1}(t,t')$ can be obtained analogously to Ref. 26, by replacing the integral in Eq. (2.21a) by a sum and then making use of $T_0^{-1}(t,t')T_0(t,t')=I$, and finally again replacing the sum by an integral:

$$T_0^{-1}(t,t')=\overline{\mathcal{T}}\exp\left(i\int_{t'}^t dt_1Q(t_1)L_0\right), \qquad (2.21b)$$

where $\overline{\mathcal{T}}$ is the reverse time-ordering operator and $L_0\equiv(1/\hbar)[H_0,\ldots]$, $L_{SR}\equiv(1/\hbar)[H_{SR},\ldots]$. This expansion of operator $T(t,t')$ in powers of the interaction is valid for all time-dependent $P(t)$ operators, which do not necessarily have to have the same form as ours.

We now let the operator $[i(\tilde{L}_S+L_{SR})\tilde{F}_n^S]$ act upon Eq. (2.16) and afterwards take the trace over it, which gives us the desired EM's for the EV's:

$$\partial\langle F_n^S\rangle_t/\partial t = i\,\mathrm{Tr}_{SR}\{[(\tilde{L}_S+L_{SR})\tilde{F}_n^S]\sigma_S(t)\otimes\rho_R(0)\} + i\,\mathrm{Tr}_{SR}\{[(\tilde{L}_S+L_{SR})\tilde{F}_n^S]T(t,0)[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\}$$
$$+\int_0^t dt'\,\mathrm{Tr}_{SR}\{[(\tilde{L}_S+L_{SR})\tilde{F}_n^S]T(t,t')[\tilde{L}_R+Q(t')(\tilde{L}_S+L_{SR})]\sigma_S(t')\otimes\rho_R(0)\}\ ,\quad n=1,\ldots,m, \qquad (2.22)$$

where we used $\mathrm{Tr}(ALB)=-\mathrm{Tr}[(LA)B]$ and $\tilde{L}_S\equiv L_S\otimes I_R$, $\tilde{L}_R\equiv I_S\otimes L_R$.

Since by Eq. (2.10), $\sigma_S(t)$ is only a function of the EV's $\langle F_n^S\rangle_t$ and according to Eqs. (2.19) and (2.13) the operators $T(t,t')$, $P(t)$ depend through $\sigma_S(t)$ also only upon the $\langle F_n^S\rangle_t$, the Eqs. (2.22) are exact closed integro-differential equations for the EV's $\langle F_n^S\rangle_t$ of the system of interest S as the only unknowns, where the variables of system R appear only as EV's taken over the initial density operator $\rho_R(0)$ of R [the initial density operators $\rho_R(0)$, $\rho_S(0)$ of R and S are assumed given]. What is especially important, is that it is always possible for a special choice of initial conditions and SO's to make

$$\rho_S(0)=\sigma_S(0) \qquad (2.23)$$

valid, so that the inhomogeneous terms which contain the difference $[\rho_S(0)-\sigma_S(0)]$ vanish.

Substituting Eqs. (2.20) and (2.21) into EM's, Eq. (2.22), we can write these equations as a series expansion in powers of the interaction H_{SR}.

It is, however, possible to make one more significant simplification in Eqs. (2.22) and (2.17), if we choose the sets for S-system operators $\{F_n^S,\ n=1,\ldots,m\}$ such that the following relation holds:

$$L_S F_n^S=\sum_{n'=1}^{m}\alpha_{nn'}F_{n'}^S\ , \qquad (2.24)$$

where $\alpha_{nn'}$ are certain coefficients. This assumption can be easily fulfilled and is also used by other authors (see Refs. 27, 28, and Ref. 18, §25.1).

Namely, by inserting Eq. (2.24) into Eqs. (2.22), we obtain

$$\partial\langle F_n^S\rangle_t/\partial t = i\sum_{n'}\alpha_{nn'}\langle F_{n'}^S\rangle_t + i\,\mathrm{Tr}_{SR}[(L_{SR}\tilde{F}_n^S)\sigma_S(t)\otimes\rho_R(0)] + i\sum_{n'}\alpha_{nn'}a_{n'}(t)+ib_n(t)$$
$$+\sum_{n'}\alpha_{nn'}\int_0^t dt'\,c_{n'}(t,t')+\int_0^t dt'\,d_n(t,t')+\int_0^t dt'\,e_n(t,t')\ ,\quad n=1,\ldots,m, \qquad (2.25)$$

where we define

$$a_{n'}(t)\equiv\mathrm{Tr}_{SR}\{\tilde{F}_{n'}^S T(t,0)[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\}\ , \qquad (2.25a)$$

$$b_n(t)\equiv\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}_n^S)T(t,0)[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\}\ , \qquad (2.25b)$$

$$c_{n'}(t,t')\equiv\mathrm{Tr}_{SR}\{\tilde{F}_{n'}^S T(t,t')[\tilde{L}_R+Q(t')(\tilde{L}_S+L_{SR})]\sigma_S(t')\otimes\rho_R(0)\}\ , \qquad (2.25c)$$

$$d_n(t,t')\equiv\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}_n^S)T(t,t')Q(t')\tilde{L}_S\sigma_S(t')\otimes\rho_R(0)\}\ , \qquad (2.25d)$$

$$e_n(t,t')\equiv\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}_n^S)T(t,t')[\tilde{L}_R+Q(t')L_{SR}]\sigma_S(t')\otimes\rho_R(0)\}\ . \qquad (2.25e)$$

Following Eq. (2.14) we can take the $P(t)$ operator into the trace of $a_{n'}(t)$ and $c_{n'}(t,t')$, and, employing equations

$$P(t)T(t,t')=P(t)\ , \qquad (2.26)$$

$$P(t)T(t,t')Q(t')=0\ , \qquad (2.27)$$

which both can be obtained through Eqs. (2.19) and (2.15), we get $a_{n'}(t)=c_{n'}(t,t')=0$. Moreover, according to Eq. (B8) in Appendix B, it results that $d_n(t,t')=0$. Further, Eq. (2.24), as we show in Appendix A, brings an essential simplification of operator $T(t,t')$ in $b_n(t)$ and $e_n(t,t')$.

Thus we finally obtain a much simpler form of the exact nonlinear coupled integro-differential equations for the m unknowns $\langle F_n^S\rangle_t$ under the condition that the SO's fulfills the relation (2.24):

$$\partial\langle F_n^S\rangle_t/\partial t = i\sum_{n'}\alpha_{nn'}\langle F_{n'}^S\rangle_t + i\,\mathrm{Tr}_{SR}[(L_{SR}\tilde{F}_n^S)\sigma_S(t)\otimes\rho_R(0)] + i\,\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}_n^S)U(t,0)e^{-itL_0}[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\}$$
$$+\int_0^t dt'\,\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}_n^S)U(t,t')e^{-i(t-t')L_0}[\tilde{L}_R+Q(t')L_{SR}]\sigma_S(t')\otimes\rho_R(0)\}\ ,\quad n=1,\ldots,m \qquad (2.28)$$

with

$$U(t,t') \equiv \mathcal{T} \exp\left(-i\int_{t'}^{t} dt_1 e^{-i(t-t_1)L_0} Q(t_1) L_{SR} e^{i(t-t_1)L_0}\right), \tag{2.29}$$

and the EM for $\sigma_S(t)$, Eq. (2.17), which is important for an eventual comparison with other papers deriving only master equations for density operators, can also be written in a simpler form:

$$[\partial\sigma_S(t)/\partial t]\otimes\rho_R(0) = -iL_S\sigma_S(t)\otimes\rho_R(0) - iP(t)L_{SR}\sigma_S(t)\otimes\rho_R(0) - iP(t)L_{SR}U(t,0)e^{-itL_0}[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)$$
$$-\int_0^t dt' P(t)L_{SR}U(t,t')e^{-i(t-t')L_0}[\tilde{L}_R + Q(t')L_{SR}]\sigma_S(t')\otimes\rho_R(0). \tag{2.30}$$

[It is obvious that the EM's for the EV's $\langle F_n^S\rangle_t$ can always be obtained from the EM for $\sigma_S(t)\otimes\rho_R(0)$.] Besides, if we choose SO's such that the special initial condition (2.23) is satisfied, then the inhomogeneous terms, which contain the difference $[\rho_S(0)-\sigma_S(0)]$, vanish in all equations derived up to now.

If $\lambda_n(t)$ could not be directly eliminated in $\sigma_S(t)$, i.e., if $\sigma_S(t)$ could not be directly expressed by the EV's $\langle F_n^S\rangle_t$ (in Refs. 29 and 30 we consider such cases where that can be done), then it is more appropriate, in addition to the unknowns $\langle F_n^S\rangle_t$, also to consider the $\lambda_n(t)$ as unknowns. By inserting Eq. (2.6) into Eqs. (2.22) or (2.28) and adding equations

$$\langle F_n^S\rangle_t = \mathrm{Tr}_S\left\{F_n^S \frac{\exp(-\sum_n \lambda_n(t)F_n^S)}{\mathrm{Tr}_S[\exp(-\sum_n\lambda_n(t)F_n^S)]}\right\},$$
$$n=1,\ldots,m, \tag{2.31}$$

we obtain $2m$ coupled integro-differential equations for $2m$ unknowns $\langle F_n^S\rangle_t$ and $\lambda_n(t)$.

Finally it is important to note that the derived EM's, Eqs. (2.17), (2.22), and (2.25), are valid not only for such a $\sigma_S(t)$ which has the form of a generalized canonical density operator, but also for all density operators, which are functions of the EV's of a given SO's: $\{F_n^S,\ n=1,\ldots,m\}$ such that $\langle F_n^S\rangle_t = \mathrm{Tr}_S[F_n^S\sigma_S(t)]$ and $\mathrm{Tr}_S\sigma_S(t)=1$ are fulfilled. [Eqs. (2.28) and (2.30) are valid only if $\sigma_S(t)$ satisfies Eq. (B8) also].

But if we want to remove the inhomogeneous terms in the EM's, i.e., to identify $\rho_S(0)$ with $\sigma_S(0)$ for special initial conditions, then we must choose such a $\sigma_S(t)$ which gives a generalized canonical density operator at the initial time $t=0$, (however, for $t>0$ this is not required). Sometimes, as we will show in Ref. 30, it is of great importance to choose such a $\sigma_S(t)$ which is only at the initial time equal to a generalized canonical density operator. Since in such cases this makes it possible to express the $\lambda_n(t)$ directly by the EV's $\langle F_n^S\rangle_t$, we thus obtain a significantly simpler form of closed EM's for the $\langle F_n^S\rangle_t$.

III. A MODIFIED ZUBAREV METHOD FOR OPEN SYSTEMS

In this section we modify the Zubarev method[15-18] which has been applied to isolated systems, so that it becomes applicable to open systems, and the analogy and differences as compared to the modified Robertson method are pointed out.

The modifying of the Zubarev method can be carried out quite analogously to that of the Robertson method in the preceding section, namely by using a generalized canonical density operator (in Zubarev's notation: quasiequilibrium density operator) $\sigma_S(t)$, defined by Eq. (2.6), with the difference that (1) the initial condition has to be taken at $t_0=-\infty$ (in the Robertson method was $t_0=0$), (2) instead of the statistical density operator $\rho(t)$ the Zubarev nonequilibrium statistical operator $\rho^{\mathrm{NSO}}(t)$, which also describes the total system $S+R$, has to be used, (3) the SO's $\{F_n^S,\ n=1,\ldots,m\}$ must always be chosen so that the initial condition (2.9) takes the following form:

$$\rho^{\mathrm{NSO}}(-\infty) = \sigma_S(-\infty)\otimes\rho_R(-\infty),\quad \mathrm{Tr}_R\rho_R(-\infty)=1. \tag{3.1}$$

The boundary condition that selects those solutions of the Liouville equation which satisfy the initial condition (3.1) is

$$\lim e^{it_1L}\rho^{\mathrm{NSO}}(t+t_1) = \lim_{t_1\to-\infty} e^{it_1L}\sigma_S(t+t_1)\otimes\rho_R(-\infty), \tag{3.2}$$

and this is equivalent to inserting infinitesimal sources into the Liouville equation for the total system $S+R$[31,32]

$$\partial\rho^{\mathrm{NSO}}(t)/\partial t + iL\rho^{\mathrm{NSO}}(t) = -\epsilon[\rho^{\mathrm{NSO}}(t) - \sigma_S(t)\otimes\rho_R(-\infty)], \tag{3.3}$$

with $\epsilon\to 0^+$ after the thermodynamical limit has been taken. The solution of Eq. (3.3) is[32]

$$\rho^{\mathrm{NSO}}(t) = \epsilon\int_{-\infty}^{0} dt' e^{\epsilon t'} e^{it'L}\sigma_S(t+t')\otimes\rho_R(-\infty). \tag{3.4}$$

The boundary condition (3.2), i.e., the infinitesimal sources in the Liouville equation (3.3) select the retarded solution of the exact Liouville equation (2.3).

Let the operator $[i(\tilde{L}_S + L_{SR})\tilde{F}_n^S]$ act on Eq. (3.4), afterwards taking the trace over it and also taking into account that

$$\langle \tilde{F}_n^S \rangle_t = \lim_{\epsilon \to 0^+} \mathrm{Tr}_{SR}[\rho^{\mathrm{NSO}}(t)\tilde{F}_n^S] = \mathrm{Tr}_S[\sigma_S(t)F_n^S] , \tag{3.5}$$

we obtain the EM's for the EV's of S-system operators:

$$\partial\langle F_n^S\rangle_t/\partial t = i\epsilon \int_{-\infty}^0 dt' e^{\epsilon t'} \mathrm{Tr}_{SR}\{[(\tilde{L}_S + L_{SR})\tilde{F}_n^S] e^{it'L}\sigma_S(t+t') \otimes \rho_R(-\infty)\}, \quad n = 1, \ldots, m \tag{3.6}$$

($\epsilon \to 0^+$ after taking the thermodynamical limit).

If for the operators F_n^S the relation (2.24) holds, then using Eq. (3.4) we can write

$$\partial\langle F_n^S\rangle_t/\partial t = i\sum_{n'} \alpha_{nn'}\langle F_{n'}^S\rangle_t + i\epsilon\int_{-\infty}^0 dt' e^{\epsilon t'} \mathrm{Tr}_{SR}\Big[(L_{SR}\tilde{F}_n^S) e^{it'L_0} \times \mathcal{T}\exp\Big(i\int_0^{t'} dt'' L_{SR}(t'')\Big)\sigma_S(t+t')\otimes\rho_R(-\infty)\Big], \quad n = 1, \ldots, m \tag{3.7}$$

with

$$L_{SR}(t'') \equiv e^{-it''L_0} L_{SR} e^{it''L_0} , \tag{3.7a}$$

where we expanded $\exp[it(L_0 + L_{SR})]$ in powers of the interaction H_{SR}, using the Dyson time-ordering operator $\mathcal{T}$.

We can also get EM's quite analogous to Eqs. (2.22) by applying the modified Robertson technique to the Liouville equation with infinitesimal sources. Namely, by use of Eq. (3.3) we obtain

$$[\partial\sigma_S(t)/\partial t] \otimes \rho_R(-\infty) = -iP(t)L\rho^{\mathrm{NSO}}(t) , \tag{3.8}$$

where the only difference between $P(t)$ appearing here and that defined by Eq. (2.13) is the replacement of $\rho_R(0)$ with $\rho_R(-\infty)$. Thus, Eq. (2.16) becomes[33]

$$\rho^{\mathrm{NSO}}(t) - \sigma_S(t)\otimes\rho_R(-\infty) = -i\int_{-\infty}^t dt' e^{\epsilon(t'-t)} T(t,t')Q(t')L\sigma_S(t')\otimes\rho_R(-\infty) . \tag{3.9}$$

[Because of the initial condition (3.1), the equation has no inhomogeneous term.]

As we can see, Eqs. (3.4) and (3.9) represent two equivalent solutions of the Liouville equation with infinitesimal sources (3.3). In the Zubarev method the equation of type (3.4) is used, while in the Robertson method the equation of type (3.9).

Let the operator $[i(\tilde{L}_S + L_{SR})\tilde{F}_n^S]$ act upon Eq. (3.9) and, carrying out the trace over it, we obtain the EM's:

$$\partial\langle F_n^S\rangle_t/\partial t = i\mathrm{Tr}_{SR}\{[(\tilde{L}_S + L_{SR})\tilde{F}_n^S]\sigma_S(t)\otimes\rho_R(-\infty)\} + \int_{-\infty}^t dt' e^{\epsilon(t'-t)}\mathrm{Tr}_{SR}\{[(\tilde{L}_S + L_{SR})\tilde{F}_n^S]T(t,t')[\tilde{L}_R + Q(t')(\tilde{L}_S + L_{SR})]\sigma_S(t')\otimes\rho_R(-\infty)\} , \quad n = 1, \ldots, m \tag{3.10}$$

($\epsilon \to 0^+$ after taking the thermodynamical limit).

Equation (3.6) and Eq. (3.10) are equivalent equations of motion for the EV's $\langle F_n^S\rangle_t$.

Consequently, we see that the EM's obtained by the modified Zubarev method differ from those of the modified Robertson method only by (1) an additional damping factor which appears in the integral, and which is the reason for the time irreversibility of the EM's and (2) using the initial time $t_0 = -\infty$ instead of $t_0 = 0$.

IV. FURTHER SIMPLIFICATION OF THE EQUATIONS OF MOTION FOR CERTAIN ASSUMPTIONS REGARDING THE IRRELEVANT SYSTEM R

Now we make some usual assumptions regarding the system R:

(i) The initial density operator of R is a stationary distribution,

$$L_R\rho_R(0)=0\,.$$

In most cases this condition is satisfied for system R. This is always the case when the initial state of system R is given by an eigenstate or a statistical mixture of eigenstates of H_R, or when $\rho_R(0)$ is given as a function of H_R only.

(ii) The average value of the interaction in the initial state of R is zero,

$$\mathrm{Tr}_R[H_{SR}\rho_R(0)]=0\,,$$

i.e., the system R does not exert any external driving force on system S.

These assumptions regarding the system R are very suitable when R has the properties of a reservoir.

It follows from assumption (ii) that both

$$\mathrm{Tr}_R[(L_{SR}\tilde{F}^S_N)\sigma_S(t)\otimes\rho_R(0)]$$

and

$$P(t)L_{SR}(t)\otimes\rho_R(0)$$

vanish. Consequently, under the assumptions (i) and (ii) the EM's (2.28) and (2.30) reduce to

$$\frac{\partial\langle F^S_n\rangle_t}{\partial t}=i\sum_{n'=1}^{m}\alpha_{nn'}\langle F^S_{n'}\rangle_t+i\,\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}^S_n)U(t,0)e^{-itL_S}[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\}$$
$$+\int_0^t dt'\,\mathrm{Tr}_{SR}[(L_{SR}\tilde{F}^S_n)U(t,t')e^{-i(t-t')L_0}L_{SR}\sigma_S(t')\otimes\rho_R(0)],\quad n=1,\ldots,m \tag{4.1}$$

and

$$\frac{\partial\sigma_S(t)}{\partial t}\otimes\rho_R(0)=-iL_S\sigma_S(t)\otimes\rho_R(0)-iP(t)L_{SR}U(t,0)e^{-itL_S}[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)$$
$$-\int_0^t dt'P(t)L_{SR}U(t,t')e^{-i(t-t')L_0}L_{SR}\sigma_S(t')\otimes\rho_R(0)\,. \tag{4.2}$$

For the special initial condition (2.23), the inhomogeneous terms with $[\rho_S(0)-\sigma_S(0)]$ vanish.

Up to now, we have not made any approximation as to the strength of the interaction between the systems S and R. Equations (4.1) and (4.2) in the approximation of second order in the interaction H_{SR} become

$$\frac{\partial\langle F^S_n\rangle_t}{\partial t}=i\sum_{n'=1}^{m}\alpha_{nn'}\langle F^S_{n'}\rangle_t$$
$$+\int_0^t d\tau\,\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}^S_n)\exp[-i\tau(\tilde{L}_S+\tilde{L}_R)]L_{SR}\exp[-i(t-\tau)L_S][\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\}$$
$$+\int_0^t d\tau\,\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}^S_n)\exp[-i\tau(\tilde{L}_S+\tilde{L}_R)]L_{SR}\sigma_S(t-\tau)\otimes\rho_R(0)\},\quad n=1,\ldots,m \tag{4.3}$$

and

$$\frac{\partial\sigma_S(t)}{\partial t}\otimes\rho_R(0)=-iL_S\sigma_S(t)\otimes\rho_R(0)$$
$$-\int_0^t d\tau P(t)L_{SR}\exp[-i\tau(\tilde{L}_S+\tilde{L}_R)]L_{SR}\exp[-i(t-\tau)L_S][\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)$$
$$-\int_0^t d\tau P(t)L_{SR}\exp[-i\tau(\tilde{L}_S+\tilde{L}_R)]L_{SR}\sigma_S(t-\tau)\otimes\rho_R(0)\,, \tag{4.4}$$

where we used the relations

$$\mathrm{Tr}_{SR}\{(L_{SR}\tilde{F}^S_n)e^{-itL_0}[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\}=0$$

and

$$P(t_1)L_{SR}e^{-it_1L_0}[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)=0\,,$$

which both follow from assumptions (i) and (ii).

Since according to assumption (ii) the approximation of the first order in the interaction H_{SR} vanishes, the approximation of the second order is the lowest order of the approximation, the so-called Born approximation. This Born approximation is usually good if the SO's is chosen big enough, the initial condition (2.23) is fulfilled (i.e., the inhomogeneous terms vanish), and the system R is a reservoir.

With the modified Zubarev method we obtain quite analogous equations if the special initial condition (3.1) is satisfied and the assumptions (i) and (ii) are made at $t_0 = -\infty$ (see Sec. III).

V. DISCUSSION

In the preceding sections we obtained a set of exact closed EM's for the system of interest S, in which the variables of the system R appear only in the form of EV's taken over the initial state of R. These equations are non-Markovian. The reduction to a Markovian form will be considered in our subsequent papers (see Refs. 29 and 30).

This formalism is attractive because we always obtain closed EM's for the EV's of a set of S-system operators. We have expressed these equations as a power series in the interaction between S and R. But we can truncate this expansion at low order only if the state of the irrelevant system R never deviates appreciably from the initial state of R. This is always the case when R is a large system in equilibrium (reservoir), which interacts so weakly with a small system S that the equilibrium of system R is hardly disturbed. Otherwise a partial resummation of the perturbative expansion should be made.

Moreover, the quality of the approximation is dependent on the number of operators included in the set. The greater the number of linearly independent operators in the set, the better the approximation in the truncation of the series. (More will be discussed about this in Refs. 29 and 30, where the application of this method to the interaction of N two-level atoms with the radiation field will be treated.)

The advantage of the modified Robertson method is that the projector used directly picks out the EV's that are of interest and thereby gives closed EM's for them. (In case of the modified Zubarev method this is obtained without projectors, by inserting infinitesimal sources into the Liouville equation.) Construction of such projectors in methods using time-independent projectors is not usually possible.

Although $\sigma_S(t)$ determines only the EV's of S-system operators included in a given set and gives no information about the EV's of the remaining S-system operators, these EV's can be determined from the connecting equation between $\rho(t)$ and $\sigma_S(t) \otimes \rho_R(0)$ [or $\rho^{\rm NSO}(t)$ and $\sigma_S(t) \otimes \rho_R(-\infty)$ in the Zubarev method], when the EV's of operators contained in the given set are already calculated from the EM's.

Moreover, the equations obtained by the modified Robertson method contain inhomogeneous terms which can easily be made to vanish through special initial conditions. (In the Zubarev method this simplification is made *a priori*, since the obtained equations are valid only for such special initial conditions.) Such a simplification for special initial conditions is not possible in the case of methods using the Argyres-Kelley projector,[1-7] because no such inhomogeneous terms exist.

Finally, we would like to mention once again that as long as no special properties of canonical density operators are used, the EM's obtained by the modified Robertson method are valid for every density operator $\sigma_S(t)$ which is a function of the EV's $\langle F_n^S \rangle_t$, $n = 1, \ldots, m$ so that $\langle F_n^S \rangle_t = \mathrm{Tr}_S[F_n^S \sigma_S(t)]$ and $\mathrm{Tr}_S \sigma_S(t) = 1$ are fulfilled. [In addition to this, in the modified Zubarev method Eq. (3.1) must also be satisfied.] Consequently, the $\sigma_S(t)$ appearing in the obtained EM's is not restricted to the generalized canonical density operators.

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APPENDIX A

By using the relation (2.24) we obtain an essential simplification for the operator $T(t,t')$ in Eqs. (2.25), namely, it follows that

$$P(t)L_0^k[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0) = \sum_{n,n',\ldots,n^{(k)}} (-\alpha_{nn'})\cdots(-\alpha_{n^{(k-1)}n^{(k)}})\rho_R(0)\otimes\frac{\partial\sigma_S(t)}{\partial\langle F_n^S\rangle_t} \times \mathrm{Tr}_{SR}\{\tilde{F}^S_{n^{(k)}}[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\} = 0\,, \quad k=0,1,2,\ldots, \tag{A1}$$

where we used Eq. (2.13). Employing Eq. (A1) and Eqs. (2.21a) and (2.21b) we get

$$T_0^{\pm 1}(t,0)[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0) = e^{\mp itL_0}[\rho_S(0)-\sigma_S(0)]\otimes\rho_R(0)\,, \tag{A2}$$

i.e., all the terms containing the $P(t)$ operator vanish. Moreover, according to Eqs. (2.24) and (2.14) for an arbitrary operator A in $\mathcal{H}_S \otimes \mathcal{H}_R$, it holds that

$$P(t)L_0^k[I-P(t')]A = \sum_{n,n',\ldots,n^{(k)}} (-\alpha_{nn'}) \cdots (-\alpha_{n^{(k-1)}n^{(k)}})\rho_R(0) \otimes \frac{\partial \sigma_S(t)}{\partial \langle F_n^S \rangle_t} \{\mathrm{Tr}_{SR}(\tilde{F}^S_{n^{(k)}}A) - \mathrm{Tr}_{SR}[\tilde{F}^S_{n^{(k)}}P(t')A]\} = 0, \quad k=0,1,2\ldots \quad \text{(A3)}$$

And from this follows that

$$T_0^{\pm 1}(t,t')[I-P(t')]A = e^{\mp i(t-t')L_0}[I-P(t')]A. \quad \text{(A4)}$$

It holds also that

$$P(t)L_0^k \tilde{L}_R A = 0, \quad k=0,1,2,\ldots \quad \text{(A5)}$$

and therefore

$$T_0^{\pm 1}(t,t')\tilde{L}_R A = e^{\mp i(t-t')L_0}\tilde{L}_R A. \quad \text{(A6)}$$

APPENDIX B

The proof that $d_n(t,t')$ defined by Eq. (2.25) vanishes is as follows:

By using Eq. (31) in Ref. 13, we can write

$$L_S\sigma_S(t) = -\sum_n \lambda_n(t) \int_0^1 dx \sigma_S(t)^x (L_S F_n^S)\sigma_S(t)^{1-x}$$
$$= -\sum_{n,n'} \lambda_n(t)\alpha_{nn'} \int_0^1 dx \sigma_S(t)^x F_{n'}^S \sigma_S(t)^{1-x}, \quad \text{(B1)}$$

where we used Eq. (2.24). Further, taking into account Eq. (2.6), we have

$$\sum_{n,n'} \lambda_n(t)\alpha_{nn'}\langle F_{n'}^S\rangle_t = 0. \quad \text{(B2)}$$

Then by using Eq. (B2) and the definition (6) in Ref. 14,

$$\overline{F}_{n'}^S \equiv \int_0^1 \sigma_S(t)^x F_{n'}^S \sigma_S(t)^{-x} dx - \langle F_{n'}^S\rangle_t, \quad \text{(B3)}$$

we can write Eq. (B1) as follows:

$$L_S\sigma_S(t) = -\sum_{n,n'} \lambda_n(t)\alpha_{nn'}\overline{F}_{n'}^S\sigma_S(t). \quad \text{(B4)}$$

Now we define a new operator $P^S(t)$ which is used by Robertson in Refs. 13 and 14 in a quite analogous form:

$$P^S(t)A^S \equiv \sum_n \frac{\partial \sigma_S(t)}{\partial \langle F_n^S\rangle_t} \mathrm{Tr}_S(F_n^S A^S), \quad \text{(B5)}$$

where A^S is any operator acting in Hilbert space $\mathcal{H}_S$.

Then, by using Eq. (A7) in Ref. 14, we obtain

$$P^S(t)\overline{F}_{n'}^S\sigma_S(t) = \overline{F}_{n'}^S\sigma_S(t). \quad \text{(B6)}$$

From this equation follows that

$$P(t)\overline{F}_{n'}^S\sigma_S(t) \otimes \rho_R(0) = \overline{F}_{n'}^S\sigma_S(t) \otimes \rho_R(0), \quad \text{(B7)}$$

and with Eq. (B4) we obtain

$$P(t)L_S\sigma_S(t) \otimes \rho_R(0) = L_S\sigma_S(t) \otimes \rho_R(0). \quad \text{(B8)}$$

According to this equation, $d_{nn'}(t)$ vanishes.

APPENDIX C

We will now make a Kawasaki-Gunton modification[23,24] of our projection operator $P(t)$ [defined by Eq. (2.13)] to obtain a formalism which more closely resembles the methods using Zwanzig's projection technique to derive master equations for the relevant parts of the density operator $\rho(t)$.[1-12] The modified projection operator $\overline{P}(t)$ can be written as follows:

$$\overline{P}(t)A = \left(\sigma_S(t) - \sum_n \frac{\partial \sigma_S(t)}{\partial \langle F_n^S\rangle_t}\langle F_n^S\rangle_t\right) \otimes \rho_R(0)\mathrm{Tr}_{SR}A$$
$$+ \sum_n \frac{\partial \sigma_S(t)}{\partial \langle F_n^S\rangle_t} \otimes \rho_R(0)\mathrm{Tr}_{SR}(F_n^S A), \quad \text{(C1)}$$

where A is an arbitrary operator in $\mathcal{H}_S \otimes \mathcal{H}_R$ and the last term of the operator $\overline{P}(t)$ is identical to the earlier projection operator $P(t)$. $\overline{P}(t)$ has the following properties:

$$\overline{P}(t)\rho(t) = \sigma_S(t) \otimes \rho_R(0), \quad \text{(C2)}$$

$$\overline{P}(t)\frac{\partial \rho(t)}{\partial t} = \frac{\partial \sigma_S(t)}{\partial t} \otimes \rho_R(0), \quad \text{(C3)}$$

$$\overline{P}(t)\overline{P}(t')A = \overline{P}(t)A, \quad \text{(C4)}$$

$$\mathrm{Tr}_{SR}[\tilde{F}_n^S\overline{P}(t)A] = \mathrm{Tr}_{SR}(\tilde{F}_n^S A), \quad \text{(C5)}$$

$$\overline{P}(t)LA = P(t)LA. \quad \text{(C6)}$$

Consequently, the new projection operator $\overline{P}(t)$ picks out the relevant part of the density operator $\rho(t)$ as the projection operators do in Refs. 1–12 and moreover has all the properties of our earlier operator $P(t)$; i.e., in all the equations derived in this paper the operator $P(t)$ can be replaced by the modified operator $\overline{P}(t)$.

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