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The Time-Local View of Nonequilibrium Statistical Mechanics. I. Linear Theory of Transport and Relaxation

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The various approaches to nonequilibrium statistical mechanics may be subdivided into convolution and convolutionless (time-local) ones. While the former, put forward by Zwanzig, Mori, and others, are used most commonly, the latter are less well developed, but have proven very useful in recent applications. The aim of the present series of papers is to develop the time-local picture (TLP) of nonequilibrium statistical mechanics on a new footing and to consider its physical implications for topics such as the formulation of irreversible thermodynamics. The most natural approach to TLP is seen to derive from the Fourier–Laplace transform $\tilde{C}(z)$ of pertinent time correlation functions, which on the physical sheet typically displays an essential singularity at $z = \infty$ and a number of macroscopic and microscopic poles in the lower half-plane corresponding to long- and short-lived modes, respectively, the former giving rise to the autonomous macrodynamics, whereas the latter are interpreted as doorway modes mediating the transfer of information from relevant to irrelevant channels. Possible implications of this doorway mode concept for so-called extended irreversible thermodynamics are briefly discussed. The pole structure is used for deriving new kinds of generalized Green–Kubo relations expressing macroscopic quantities, transport coefficients, e.g., by contour integrals over current–current correlation functions obeying Hamiltonian dynamics, the contour integration replacing projection. The conventional Green–Kubo relations valid for conserved quantities only are rederived for illustration. Moreover, $\tilde{C}(z)$ may be expressed by a Laurent series expansion in positive and negative powers of z, from which a rigorous, general, and straightforward method is developed for extracting all macroscopic quantities from so-called secularly divergent expansions of $\tilde{C}(z)$ as obtained from the application of conventional many-body techniques to the calculation of $\tilde{C}(z)$.

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The expressions are formulated as time scale expansions, which should rapidly converge if macroscopic and microscopic time scales are sufficiently well separated, i.e., if lifetime ("memory") effects are not too large.

KEY WORDS: Statistical mechanics; Liouville equation; irreversible processes; correlation functions; Green–Kubo relations; transport coefficients; non-Markovian processes.

1. INTRODUCTION

From experience, we know that the relaxation toward equilibrium of a closed macroscopic system held at nonequilibrium at some initial instant of time t_0 is well described by phenomenological relaxation or transport equations. Ready examples are the equtions of Boltzmann or Enskog in kinetic theory, various master equations, the hydrodynamic equations, Fick's and Fourier's laws, and the equations of chemical kinetics. Equations of this kind, called macroscopic evolution equations (MEE) in the following, provide us with a reduced or contracted description, which is closed, at the phenomenological level at least, of macroscopic systems in terms of just a few observables (the five hydrodynamic fields, for example) instead of the overwhelmingly large number of microscopic degrees of freedom.

Ever since Boltzmann, nonequilibrium statistical mechanics (NESM) has strived after deriving and generalizing the equations of macrophysics from those of microphysics in order both to define precisely and extend conveniently the region of validity of those MEE and also for revealing the origins of their (approximate) validity. Moreover, NESM has also attempted to clarify the role of fluctuations in the relaxation process, to provide a microscopic foundation of irreversible thermodynamics, and, as an ultimate goal, to elucidate the connection between the reversible and deterministic laws of physics and the second law of thermodynamics.

Many approaches have been developed for solving these tasks, the one of most elegance and common use certainly being the Mori–Zwanzig method[1–4] and generalizations[5–7] thereof based on the introduction of projection operators into the theory. This way, one obtains a systematic method for eliminating the so-called irrelevant variables in favor of the relevant ones, which are the observables of physical interest. Close to equilibrium, i.e., in the linear case, NESM based on this approach acquires a very satisfying, essentially closed form, which is characterized by the following features:

1. The MEE are nonlocal in time, i.e., they are obtained as first order Volterra *integrodifferential* equations, which is manifest by their containing

a time convolution. This may be viewed as a direct consequence of causality and is usually interpreted as a memory, so that the phenomenological MEE, which are autonomous (memory-free) equations, are said to be generalized. As a consequence, the phenomenological transport kernels[2] are replaced with "frequency"-dependent ones.

2. The latter are given explicitly by generalized Green–Kubo relations relating them to correlation functions of microscopic fluxes formulated in terms of a projected Hamiltonian dynamics.

3. In the Heisenberg picture[4,6] one obtains a generalized Langevin equation describing the time evolution of observables, including their fluctuations. There exists a fluctuation-dissipation theorem of the second kind relating the random forces to the generalized transport kernels.

4. Irreversible thermodynamics is generalized in the sense that the fluxes are expressed in terms of the whole history of the forces. As in rational thermodynamics,[8] the usual notion of the thermodynamic state of the system has to be modified by including this history.

All of these features are a direct consequence of the time convolution contained in the MEE, so that in the following we will call the above approach the convolution picture (CP) of NESM. Besides the many attractive aspects of the CP, there are also some serious drawbacks. In fact, the projection operators are not uniquely defined, which may lead to ambiguities in approximations. This is particularly true in far from equilibrium (nonlinear) situations, where the projection method gets rather complicated. Moreover, the pertinent kernels are not given in terms of Hamiltonian dynamics (see point 2 above), which renders more difficult the application of familiar many-body techniques in their evaluation. For the same reason, the generalized Green–Kubo relations do not relate the transport kernels directly to measurable quantities such as ordinary time correlation functions.

There is a different approach[9–29] to NESM, called the convolutionless or time-local picture (TLP) in the following, leading to equally closed, exact MEE and generalized Langevin equations, which are first-order *differential* equations with respect to time. In general, these equations are nonautonomous ones, i.e., they also contain a memory, which manifests itself in the time dependence of the corresponding generalized transport kernels (see Section 2 for details).

[2] Since the notion of the MEE is used for a wide variety of equations, we introduce the notion of the transport kernel to denote quantities such as ordinary transport coefficients, collision or master operators, and the like.

There are mainly two routes for developing the TLP approach; one[11 20] is more concerned with the general theory also being based on the projection operator method. Typically, one starts from the CP and carries out a procedure called memory renormalization[14-16] to end up with the TLP. This is also the route followed by Balescu in kinetic theory (see §17.2 in Ref. 30).

Another route,[23-29] concerned more with applications, is based on the observation that there are typically two ansatzes used in calculating time correlation functions. One, called the inversion or memory function method, starts from the Laplace transform of the correlation function and expresses this as the inverse of something, which is essentially the memory kernel[3] or the matrix of "frequency"-dependent transport kernels. This corresponds to the CP. The other one, corresponding to the TLP, is called the exponential or cumulant expansion method and consists in starting directly from the correlation function in time representation, which gets expressed as the exponential of something that is directly related to the matrix of the time-dependent generalized transport kernels of the TLP. This method has been applied to a variety of problems, ranging from line shape problems[27-29] to the study of systems with disorder.[26,31] Although both methods lead to identical results in principle, they do not do so in approximations and the general result of the examples treated is that in approximations the TLP is superior to the CP in many cases.

Similar conclusions can also be drawn in problems closely related to NESM, such as obtaining reduced descriptions (MEE) in deterministic systems exhibiting chaos and in deriving generalized Fokker–Planck equations for stochastic differential equations of Langevin type with multiplicative colored noise. Projection operator methods of both the convolution[32] and convolutionless[33,34] type and in particular the cumulant expansion method[35,36] corresponding to the TLP are quite common there, the latter having been advocated mainly by Fox[37,38] (see also Ref. 39). The main achievement here is that the kernel of the generalized Fokker–Planck equation is obtained as a systematic series expansion in powers of the correlation time t_c of the noise, the coefficients being given explicitly in terms of ordered operator cumulants.

Actually, the expansion parameter is not t_c itself, but instead the ratio between the time scales given by t_c and the overall relaxation time t_R. Therefore, we will call such expansions *time scale expansions* (TSE). The great advantage of TSE is that in practical cases they are rapidly converging, since microscopic and macroscopic time scales usually are well separated.

[3] In field theory this is just the self-energy or mass operator.

In NESM, the TLP is less advanced. The existing MEE and generalized Langevin equations obtained by the TLP projection operator methods[11–20] are rather involved and share in an even more serious way the drawbacks of the projection method mentioned above, particularly in far from equilibrium situations. Yet there exist systematic expressions for the kernels of the MEE in the form of ordered cumulant expansions corresponding to a series expansion in powers of a small parameter, usually the density or the interaction strength. In many systems of interest, however, either there is no small physical parameter at all or the series have to be summed at least partially up to infinite order, which is not simple in the existing cumulant expansions. Therefore, it is desirable, and will be done in the present paper, to develop a TSE for NESM, too, since the corresponding expansion parameter $\xi = t_c/t_R$ is always small in realistic situations independently of the density or the interaction strength.

These, however, are technicalities, the question of using CP or TLP being more deeply rooted than just in, for example, working in time or frequency representation of the correlation functions in order to get optimized expansions. The point of main interest is that under rather general conditions CP is fully consistent with the existence of an *autonomous macrodynamics* at sufficiently late (macroscopic) times, memory effects playing a role only during an initial (microscopic) period of time (see Section 2 for details).

Now, the very existence of an autonomous macrodynamics also should have a number of important physical consequences for the theory of fluctuation and dissipation and the construction of irreversible thermodynamics, which cannot be read off so easily from the CP. Instead, the TLP provides in many respects a different *view* of NESM and allows one to look at many of its subjects under a different angle, some of the most pronounced features of which are as follows.

1. In the TLP approach the existence of an autonomous macrodynamics is most clearly exhibited in that at macroscopic times the exact MEE become autonomous, with transport kernels independent of time or frequency.[4] This is to be contrasted with point 1 above and is discussed further in Section 2.

2. Generalized Green–Kubo relations can be derived (see Section 4.1) expressing the transport kernels of the autonomous MEE and the parameters of irreversible thermodynamics in terms of the pysical (i.e., non-

[4] It is to be noted that the TLP transport kernels fully contain all of the so called non-Markovian or memory effects of the CP and therefore sometimes are called memory renormalized.

projected) time correlation functions, including exactly the so-called memory or non-Markovian effects.

3. The generalized Langevin equation can be formulated (see forthcoming paper) consisting of an *autonomous* deterministic part and a random force corresponding to stationary colored noise, which, however, is not orthogonal to the observables, the orthogonality observed in the CP obtaining only in the white noise limit (complete separation of time scales).

4. Irreversible thermodynamics is reobtained in its classical form with respect to the flux–force relation and the notion of the state, the conjugate thermodynamic forces figuring as memory renormalized ones.

Astonishingly, only a few of these findings have been obtained before, so that we think it worthwhile to devote this and the following papers to a more or less complete development of the time-local view of NESM and irreversible thermodynamics for both the linear and nonlinear theory, taking full account of so-called non-Markovian or memory effects.

The present paper is concerned with linear theory only and mainly covers points 1 and 2 of the above program. In Section 2, we present some basic relations between the CP and TLP approaches, which partly were given earlier.[40,41] Then, in Section 3 we study the structure of the Fourier–Laplace transform of the pertinent correlation functions on the physical sheet of its Riemannian surface, which, we feel, is the most natural basis for the TLP approach to NESM (see Section 3.4 for a discussion).

Having established the analytical structure of the correlation functions on the physical sheet, we then in Section 4.1 derive the generalized Green–Kubo relations announced above and by means of this obtain the TSE of the macroscopic parameters of the theory (see Section 5). These are valid in "frequency" representation, corresponding expressions in time language being given in Section 6. A short summary and some concluding remarks are given in Section 7.

2. MACROSCOPIC EVOLUTION EQUATIONS IN LINEAR THEORY

Let us consider a closed system of N particles described macroscopically by a set of observables $A_1,..., A_n$ obeying the equations of motion (Heisenberg picture)

$$\dot{A}_k(t) = iLA_k(t), \qquad k = 1,..., n \tag{2.1}$$

where the Liouvillean L is defined by

$$L... = i\{H,...\} \qquad \text{or} \qquad L... = (1/\hbar)\{H,...\} \tag{2.2}$$

H denoting the Hamiltonian of the system, $\{\cdot\}$ the Poisson bracket, and $[\cdot]$ the commutator for the classical and quantum mechanical cases, respectively. The system is assumed to be in a nonequilibrium state given by an ensemble $\rho(t_0)$ at an arbitrary time t_0 for which it is convenient to choose (cf. Ref. 6)

$$\rho(t_0) = Z^{-1}[\exp(-\beta H) - \lambda \cdot A], \qquad \lambda \cdot A = \sum_{k=1}^{n} \lambda_k A_k \tag{2.3}$$

Z being the normalization factor.

From the macroscopic point of view we are interested in the time evolution of the ensemble averages $a_k(t)$ of the observables $A_k(t)$, $k = 1,..., n$, with respect to $\rho(t_0)$. Assuming that $\rho(t_0)$ describes a situation sufficiently close to equilibrium, one obtains from linearizing Eq. (2.3) and proposing that $A_k \forall k$ is conveniently defined so that its equilibrium average is zero,

$$a(t) = -C(t - t_0)\,\lambda(t_0) \tag{2.4}$$

C being the matrix of correlation functions or correlation matrix

$$C(s) = \langle A(s) \mid A^+ \rangle = \langle A \mid e^{-iLs} \mid A^+ \rangle, \qquad s = t - t_0 \tag{2.5}$$

We introduced Liouville space notation,[42] the scalar product between any two quantities B, C being defined as

$$\langle B \mid C \rangle = \frac{1}{N!} \int d1 \cdots dN\, BC^* \rho_0$$

and

$$\langle B \mid C \rangle = \mathrm{Tr} \int_0^1 dx\, B \rho_0^x C^* \rho_0^{1-x}$$

in classical and quantum mechanics, respectively, where B, $C = B$, $C(1,..., N)$ are phase space functions in the former and operators in the latter case.

Note that $C(s)$ as given by Eq. (2.5) is an $n \times n$ matrix. This means that we always have to consider A as a column and A^+ as the row matrix $A^+ = \{A_1^*,..., A_n^*\}$, so that A^+ is the Hermitian conjugate of A. With this definition we find that the variance matrix of the equilibrium fluctuations

$$F := C(0) = \langle A \mid A^+ \rangle \tag{2.6}$$

is always a Hermitian matrix. In terms of F we have

$$a = -F\lambda \tag{2.7}$$

so that

$$a(t) = +C(t-t_0)\, F^{-1} a(t_0) \tag{2.8}$$

2.1. Evolution Equations in CP and TLP

As seen from Eq. (2.8), the evolution in time of the ensemble averages $a(t)$ is completely specified in terms of $C(t-t_0)$. Hence, replacing $t-t_0$ with t, we may restrict ourselves to the study of $C(t)$. *Instantaneous or local in time evolution equations* for $C(t)$ are readily obtained as

$$\dot{C}(t) = -I(t)\, C(t) \tag{2.9}$$

where

$$I(t) = -\dot{C}(t)\, C^{-1}(t) \tag{2.10}$$

Introducing the frequency matrix Ω,

$$\Omega = \langle A |\, L \,| A^+ \rangle\, F^{-1} = i\dot{C}(0)\, F^{-1} \tag{2.11}$$

we may rewrite $I(t)$ as

$$I(t) = i\Omega + I^{\mathrm{ir}}(t) \tag{2.12}$$

where (cf. Ref. 24)

$$I^{\mathrm{ir}}(t) = -\int_0^t dt'\, \langle \dot{A}^{\neq}(t') |\, \dot{A}^+ \rangle\, C^{-1}(t') \tag{2.13}$$

and

$$\dot{A}^{\neq}(t) = i[L - iI(t)]\, A(t)$$

Equation (2.9) represents a set of n, in general nonautonomous, differential equations. On the other hand, the Mori–Zwanzig theory yields the retarded evolution equations given by the set of integrodifferential equations

$$\begin{aligned} \dot{C}(t) &= -\int_0^t dt'\, M(t')\, C(t-t') \\ &= -i\Omega C(t) - \int_0^t dt'\, M^{\mathrm{ir}}(t')\, C(t-t') \end{aligned} \tag{2.14}$$

where

$$M^{\mathrm{ir}}(t) = \langle \dot{A}|\, Qe^{-iQLQt}Q\, |\dot{A}^{+}\rangle\, F^{-1} \tag{2.15}$$

$$Q = 1 - P, \qquad P = |A^{+}\rangle\, F^{-1}\langle A| \tag{2.16}$$

In the present paper we investigate the above evolution equations for systems where two (or several) different time scales t_R, t_c exist,

$$t_R^{-1} = \int_0^{\infty} dt\, \|M(t)\| \tag{2.17a}$$

$$t_c = t_R \int_0^{\infty} dt\, t\, \|M(t)\| \tag{2.17b}$$

where

$$\xi = t_c/t_R = \int_0^{\infty} dt\, t\, \|M(t)\| \tag{2.17c}$$

is not too large, and $M(t)$ decays sufficiently rapid, namely

$$M(t) = 0, \qquad t \gg t_c \tag{2.18}$$

Note that Eq. (2.18) is a symbolic notation that excludes algebraic decay, which has no time scale. This might seem a severe restriction, since in many cases an algebraic decay is the rule. However, as briefly sketched in Section 5.3, most of the results of the present paper can be modified to cover the latter case.

Hence, we restrict the present paper to studying the nonalgebraic case, defined by Eq. (2.18).

We note that both Eqs. (2.9) and (2.14) are exact. They are reversible and yield a decaying solution for both $t \to +\infty$ and $t \to -\infty$ (cf. Fig. 1). Moreover, both equations are nonautonomous, since time t appears explicitly, and hence display a memory behavior.

For discussing this point it is useful to consider the TLP equation for $a(t)$, i.e.,

$$\dot{a}(t) = -I(t - t_0)\, a(t) \tag{2.19}$$

which corresponds to Eq. (2.9). It is customary to call $u = t - t_0$ the age of the state, because u is the time elapsed since the initial preparation of the system. Then Eq. (2.19) shows explicitly that knowing $a(t)$ at an instant of time t is not sufficient for determining the future evolution of $a(t)$, since I

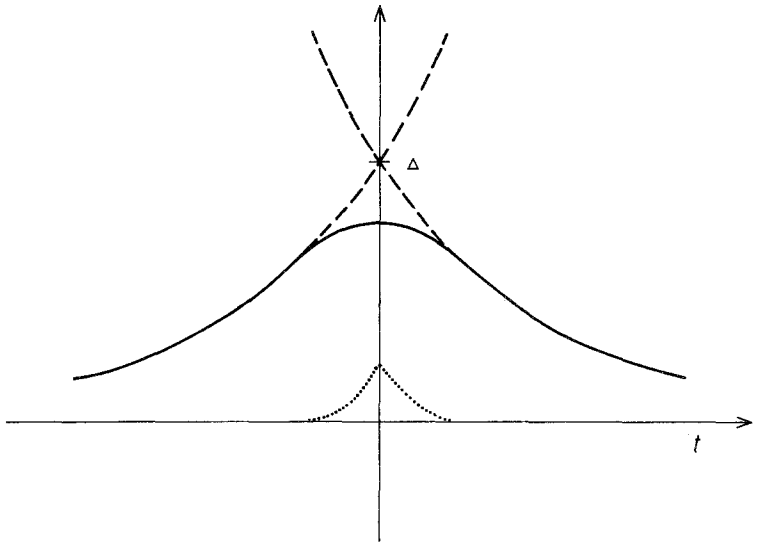

Fig. 1. Schematic representation of (—) the correlation function $C(t)$, (– –) its macroscopic parts $C^{(+)}(t)$ and $C^{(-)}(t)$, and (· · ·) the microscopic part $-C^{(m)}(t)$ representing the initial slip. The case of a single observable is considered, where $\Delta^{(+)}$ is a scalar.

still depends on the age u. Thus, one may say that the evolution of $a(t)$ is still influenced by the initial preparation of the system, i.e., we observe a memory behavior at these times.

2.2. Autonomous Macrodynamics in TLP

Under certain physical conditions (see below) it may well happen that this memory fades away with increasing age u of the system. This means that

$$\lim_{t \to \pm\infty} I(t) =: I^{(\pm)} \tag{2.20}$$

exists. $I^{(+)}$ determines the decay at macroscopic times if only $I(t)$ reaches its plateau value $I^{(+)}$ after some microscopic time t_c, i.e., symbolically,

$$I(t) = I^{(+)}, \qquad t \gg t_c \tag{2.21}$$

and we note that in general $I(t)$ but not $I^{(+)}$ depends on the initial preparation of the system.

For investigating the consequences of Eq. (2.21) we introduce $C^{(+)}$ ($C^{(-)}$), which is the solution of the autonomous equation

$$\dot{C}^{(\pm)}(t) = -I^{(\pm)} C^{(\pm)}(t) \tag{2.22}$$

for all times $-\infty < t < \infty$, and split

$$\begin{aligned} C(t) &= C^{(+)}(t) + C^{(m)}(t), \qquad t > 0 \\ C(t) &= C^{(-)}(t) + C^{(m)}(t), \qquad t < 0 \end{aligned} \tag{2.23}$$

Then, choosing the initial condition so that $C^{(+)}$ fits the wing of $C(t)$ for $t \to \infty$, i.e.,

$$C^{(\pm)}(0) =: \Delta^{(\pm)} = \lim_{t \to +\infty} [\exp(I^{(\pm)}t)]\, C(t) \tag{2.24}$$

and hence

$$C^{(\pm)}(t) = [\exp(-I^{(\pm)}t)]\, \Delta^{(\pm)} \tag{2.25}$$

we find as a consequence of Eq. (2.21)

$$C^{(m)}(t) = 0, \qquad |t| \gg t_c \tag{2.26}$$

Consequently, we may call $C^{(\pm)}$ the macroscopic branches of the correlation matrix $C(t)$, where $C^{(m)}$ represents the initial slip containing the decay of transients (cf. Fig. 1).

As a consequence of Eqs. (2.23), (2.26), and (2.8) we also note that in a sufficiently aged state the time evolution is completely determined by the macroscopic branch $C^{(+)}$ of C, i.e., we write

$$a = a^{(+)} + a^{(m)} \tag{2.27a}$$

$$a^{(+)}(t) = C^{(+)}(t - t_0)\, F^{-1} a(t_0) \tag{2.27b}$$

so that $a^{(+)}$ obeys the autonomous equation

$$\dot{a}^{(+)} = -I^{(+)} a^{(+)} \tag{2.27c}$$

and

$$a(t) = a^{(+)}(t), \qquad t - t_0 \gg t_c$$

Equations (2.22) and (2.27c) define an autonomous macrodynamics, which obviously is characterized by the fact that the rate of change $\dot{a}^{(+)}(t)$ $[\dot{C}^{(+)}(t)]$ is related to $a^{(+)}(t)$ $[C^{(+)}(t)]$ via a quantity $I^{(+)}$ that is independent on the history of the system or the age of the state considered. Hence, Eqs. (2.22) and (2.27c) are memory-free equations.

2.3. Autonomous Macrodynamics in CP

The predominance of the autonomous macrodynamics (AM) at late times as expressed by Eqs. (2.23), (2.26), or (2.27) is an immediate consequence of the plateau property (2.21) of $I(t)$. This was simply postulated and might seem rather artificial in view of the retardation contained in the CP equations (2.14) at all times.

This point is elucidated best by studying directly the solution of Eq. (2.14). As shown in Section 3, under the conditions formulated with Eqs. (2.17) and (2.18) the solution of Eq. (2.14) may be written as in Eq. (2.23), i.e., $C(t)$ is decomposed into $C^{(+)}$ and $C^{(m)}$ corresponding to the AM and the decay of transients, respectively.

In TLP the transition from the memory-retaining behavior at small t to the AM prevailing at large t is expressed by Eq. (2.21). In CP this transition is reflected more implicitly by the fact that Eq. (2.14), which is of Volterra type, changes for large t into the autonomous integrodifferential equation obeyed by $C^{(+)}$

$$\dot{C}^{(+)}(t) = -\int_0^\infty dt'\, M(t')\, C^{(+)}(t-t') \tag{2.28}$$

which is of Fredholm type.

Equations (2.25) and (2.28) are the actual macroscopic evolution equations of TLP and CP, respectively, since they describe the relaxation of the system at all *macroscopic* times. The necessary initial condition for both equations is furnished by Eq. (2.24), so that $C^{(+)}(t)$ fits the wing of $C(t)$ for $t \to \infty$. The connection between the two pictures is established by the nonlinear integral equation

$$I^{(+)} = \int_0^\infty dt\, M(t) \exp(I^{(+)} t) \tag{2.29}$$

which is obtained by introducing Eq. (2.25) into Eq. (2.28). Thus, knowing $M(t)$, one may determine $I^{(+)}$, which together with $\Delta^{(+)}$ is the only quantity of macroscopic interest.

2.4. Remarks on CP and TLP

Equations (2.22) and (2.28) may be considered as, respectively, the instantaneous and retarded representation of the same behavior, namely the simple exponential decay given by Eq. (2.25). Thus, from the macroscopic point of view, $M(t)$ may be considered as an intermediary quantity from which we may determine $I^{(+)}$ and $\Delta^{(+)}$, which are the only

quantities of macroscopic interest. The two different mathematical formulations (2.22) and (2.28) of the same decay law (2.25) differ in the microscopic expressions for their kernels $I^{(+)}$ and $M(t)$. Using Eq. (2.17), we find from Eq. (2.29)

$$I^{(+)} = \int_0^\infty dt\, M(t) + O(t_c/t_R) \tag{2.30}$$

so that $I^{(+)}$ is different from the so-called Markovian limit of CP by correction terms usually called memory or non-Markovian effects.

These result from the finite duration of microscopic events, so that we shall call them *lifetime effects* in the present paper.

Hence, the difference between $I^{(+)}$ and $M(t)$ consists in the contribution of lifetime effects. Typical such effects are produced by the influence of the surrounding medium on a single binary collision. In a dilute but strongly inhomogeneous system this is mainly a mean field effect contained in the Vlassov potential. These in-medium effects enter into $I^{(+)}$ and $M(t)$ in different ways. In particular, it has been shown recently[44] that $I^{(+)}$ is given by a completed binary collision taking place in the Vlassov potential, whereas such a physical interpretation is not possible for $M(t)$. This example might be generic. In fact, as we have seen, the correlation matrix $C(t)$ is composed of two physically different contributions $C^{(+)}$ and $C^{(m)}$ corresponding to the decay of macroscopic modes and of transients. The decay of $C^{(+)}$ is governed by the matrix of the physically relevant decay constants $I^{(+)}$. Consequently, the microscopic events entering $I^{(+)}$ pertain to the decay of macroscopic modes only, whereas $M(t)$ still contains the information on the decay of transients.

The connection between the two pictures is provided by Eq. (2.29), so that one can of course start out from a microscopic analysis of $M(t)$ to end up with $I^{(+)}$. An alternative approach may be based on the generalized Green–Kubo relations to be presented in Section 4, which relate the macroscopic parameters in a more direct way to the microscopic (Hamiltonian) dynamics of the system so that the calculation of the memory kernel $M(t)$ with its projected dynamics is avoided.

3. ANALYTICAL PROPERTIES OF $\tilde{C}(z)$ ON THE PHYSICAL SHEET

Let us consider the Fourier–Laplace transform of the correlation matrix $C(t)$, i.e.,

$$\tilde{C}(z) = \int_0^\infty dt\, e^{izt} C(t), \qquad \operatorname{Im} z > 0 \tag{3.1a}$$

the inverse transformation being given by

$$C(t) = \int_{\bar{C}} \frac{dz}{2\pi} e^{-izt} \tilde{C}(z) \tag{3.1b}$$

as usual, $\bar{C}$ representing a straight line parallel to the real axis, which lies above all of the singularities of $\tilde{C}(z)$.

It is customary to introduce

$$\tilde{\tilde{C}}(z) = \langle A | \, R(z) \, | A^+ \rangle \tag{3.2}$$

the resolvent $R(z)$ being

$$R(z) = i/(z - L)$$

which is defined for all z with $\operatorname{Im} z \neq 0$.

We have $\tilde{C}(z) = \tilde{\tilde{C}}(z)$ for $\operatorname{Im} z > 0$, but not so for $\operatorname{Im} z < 0$. Under suitable physical conditions $\tilde{\tilde{C}}(z)$ has a cut along the real axis and can be analytically continued beyond this cut from both below and above the real axis. The continuation of $\tilde{C}(z)$ [cf. Eq. (3.1a)] into the lower half-plane defines the physical sheet of the Riemannian surface of $\tilde{\tilde{C}}$ and we now study the properties of this $\tilde{C}(z)$ for $\operatorname{Im} z < 0$.

3.1. Analytical Continuation

The analytical continuation of the resolvent $R(z)$ is one of the major topics dealt with in the theory of subdynamics developed by the Brussels school, which is concerned mainly with providing a microscopic basis for the second law. Fortunately, for the purpose of the present paper, we may content ourselves with considering only some matrix elements of the resolvent, i.e., Eq. (3.2), corresponding to the pertinent correlation matrix $\tilde{C}(z)$, so that the most difficult problem of continuing the full resolvent is avoided.

Moreover, the still difficult problem of deriving the analytical properties of $\tilde{C}(z)$ directly from the microscopic dynamics is sidestepped here by tracing these properties back to the properties of the "memory" kernel $M(t)$, which are assumed to be known; i.e., we start from the convolution picture (CP) and model $M(t)$ by one of the conventional ansatzes (exponential, Gaussian, etc.) obeying Eq. (2.15). Although the simplest one in the present context, the exponential ansatz [see (A1)] is known to be unrealistic in the sense that it leads to divergent higher order sum rule expressions.[43] To avoid this drawback we shall assume $M(t)$ to be given by a Gaussian behavior and discuss the generality of the results in Section 3.4.

For a Gaussian behavior of $M(t)$ the transform

$$\tilde{M}(z) := \int_0^\infty dt\, e^{izt} M(t) \tag{3.3}$$

exists for all $z \neq \infty$ and is analytic for these z. Using Eqs. (2.14) and (2.6), we find for $\operatorname{Im} z > 0$

$$\tilde{C}(z) = \frac{i}{z + i\tilde{M}(z)} F \tag{3.4}$$

The analytical continuation of $\tilde{C}$ is now trivial; we obtain it by simply taking (3.4) as the definition of $\tilde{C}(z)$ valid for all $z \neq \infty$. What we are interested in are the singularities of $\tilde{C}(z)$, which all lie in the lower half-plane. We note first that $\tilde{M}(z)$ has an essential singularity at $z = \infty$. In fact, using Eq. (2.13b), we find $\tilde{M}(z) \sim (i/z)\langle \dot{A}|\, Q\, |\dot{A}^+\rangle$, $\operatorname{Im} z > 0$, $|z| \to \infty$, whereas from Eq. (3.3) we obtain $\|\tilde{M}(z)\| \to \infty$ for $\operatorname{Im} z < 0$ and $|z| \to \infty$. Now, if $\tilde{M}(z)$ has an essential singularity at $z = \infty$, so has $z + i\tilde{M}(z)$. Consequently, from Picard's theorem we may conclude that $\tilde{C}(z)$ also has an essential singularity at $z = \infty$. The existence of this essential singularity is connected with the sum rules[43] obeyed by the *Fourier* transform of $C(t)$ and is the main difference from the exponential ansatz, where this singularity is not obtained.

3.2. The Case of a Single Observable

For the discussion of further singularities of $\tilde{C}$ we first consider the case that our set A consists of a single variable only. Then, $\tilde{M}(z)$ is a scalar instead of a matrix and the singularities of $\tilde{C}$ follow from the dispersion relation

$$iz = \tilde{M}(z) \tag{3.5}$$

the solutions of which are given in the Appendix for both the exponential and the Gaussian case, the exponential ansatz being included since it can be treated explicitly and the results may be expected to carry over at least qualitatively to other ansatzes, too. It is shown that Eq. (3.5) has just two solutions z_M and z_m, where $z_M = O(1/t_R)$ and $z_m = O(1/t_c)$. Hence, we conclude that $\tilde{C}(z)$ is analytic everywhere except for $z = z_{M,m}$, where it has poles, and $z = \infty$, where it has an essential singularity. In the following we shall call the pole at z_M the *macroscopic pole*, whereas the remaining two singularities are called *microscopic* ones.

The reason for doing so is that upon introducing this result into Eq. (3.1b), we find by using the theory of residua that for sufficiently large times it is only the contribution of the macroscopic pole that survives in $C(t)$. This is the justification for splitting $C(t)$ as in Eq. (2.23) and shows that $C^{(+)}(t)$ is just the contribution of this pole, i.e. (see Fig. 2, below)

$$C^{(+)}(t) = e^{-iz_M t} r \tag{3.6a}$$

where the pole strength

$$r = [1 + iM'(z_M)]^{-1}, \qquad M' = \frac{d}{dz}\tilde{M} \tag{3.6b}$$

is essentially the residuum of $\tilde{C}(z)$ at $z = z_M$ and we are led to the identifications

$$I^{(+)} = iz_M = \tilde{M}(z_M) \tag{3.7}$$

$$\Delta^{(+)} = r \tag{3.8}$$

3.3. Several Observables

For discussing the analytical properties of $\tilde{C}(z)$ in the general case where $A = \{A_1, ..., A_n\}$ we need the spectral resolution of Eq. (3.4). For this purpose we introduce the eigenvectors $E_k(z)$ of $\tilde{M}(z)$, i.e.,

$$\tilde{M}(z)\, E_k(z) = m_k(z)\, E_k(z), \qquad k = 1, ..., n \tag{3.9}$$

$m_k(z)$ being the eigenvalues and each $E_k(z)$ is to be understood as a column matrix with matrix elements $E_k^i(z)$, $i = 1, ..., n$. Moreover, we introduce the corresponding eigenprojectors $P_k(z)$, which are n by n matrices with matrix elements

$$[P_k(z)]_{i,j} = E_k^i(z)\, E_k^j(z), \qquad i, j = 1, ..., n$$

so that

$$\tilde{M}(z)\, P_k(z) = m_k(z)\, P_k(z) \tag{3.10}$$

and

$$P_k(z)\, P_l(z) = \delta_{k,l} P_k(z) \tag{3.11}$$

if $\tilde{M}$ is a Hermitian matrix, which we shall assume in the following. Using Eqs. (3.10) and (3.11), we find the spectral resolution of $\tilde{C}(z)$ given by

$$\tilde{C}(z) = \sum_{k=1}^{n} \frac{iP_k(z)}{z + im_k(z)} \tag{3.12}$$

so that the singularities of $\tilde{C}$ are given by the solution of the n dispersion relations

$$iz = m_k(z), \qquad k = 1,..., n \tag{3.13}$$

Let us assume the $m_k(z)$ are given by Fourier–Laplace transforms of Gaussians or the like. Then, according to the Appendix and the discussion given in Section 3.2, we may conclude that Eq. (3.13) has just two solutions $z_{M,k}$ and $z_{m,k}$ for each $k = 1,..., n$. Therefore, for the special ansatz considered, we find that besides the essential singularity at $z = \infty$, $\tilde{C}(z)$ *has exactly n macroscopic and n microscopic poles*, the contribution of the former yielding [cf. Eq. (3.6a)]

$$C^{(+)}(t) = \sum_{k=1}^{n} e^{-iz_{M,k}t} \Pi_k \tag{3.14}$$

where $\Pi_k = r_k P_k(z_{M,k})$ and

$$r_k = [1 + im_k'(z_{M,k})]^{-1} \tag{3.15}$$

Π_k being essentially the residuum of $\tilde{C}(z)$ at $z = z_{M,k}$. Comparing Eq. (3.14) with Eq. (2.25), we immediately find

$$\Delta^{(+)} = \sum_{k=1}^{n} \Pi_k \tag{3.16}$$

so that $\Delta^{(+)}$ is the sum of the residua Π_k.

A similar relation for $I^{(+)}$ is not so easily established. The reason is that $\Pi_k \Pi_l \neq 0$, $k \neq l$, in general, since the eigenvectors $E_k(z_{M,k}) =: \bar{E}_k$ are not mutually orthogonal. Therefore, we have to form a biorthogonal set by introducing the new vectors $\bar{\bar{E}}_k$ which obey

$$\bar{\bar{E}}_k \cdot \bar{E}_l = 0, \qquad k \neq l$$

the dot denoting the scalar product. Now, we introduce the new projection matrices $\hat{P}_k$, where

$$[\hat{P}_k]_{i,j} = \bar{\bar{E}}_k^i \bar{E}_k^j \tag{3.17}$$

and find by multiplying both Eq. (2.25) and (3.14) by $\hat{P}_k$ from the left that

$$I^{(+)} = i \sum z_{M,k} \hat{P}_k \tag{3.18}$$

and

$$\hat{P}_k \Delta^{(+)} = r_k \hat{P}_k \tag{3.19}$$

Thus, we may say that $I^{(+)}$ is a non-Hermitian matrix with eigenvalue equations

$$I^{(+)}\bar{\bar{E}}_k = iz_{M,k}\bar{\bar{E}}_k, \qquad \bar{E}_k^T I^{(+)} = iz_{M,k}\bar{E}_k^T \tag{3.20}$$

the eigenvalues all being real. Note that we assumed $M(t)$ to be Hermitian and

$$\tilde{M}(z_{M,k})\,\bar{E}_k = iz_{M,k}\bar{E}_k \tag{3.21}$$

Thus, the main difference between the matrices $I^{(+)}$ and $\tilde{M}$ is seen to be contained in the fact that the eignevectors $\bar{E}_k$ are obtained from $\tilde{M}(z)$ for different values of $z = z_{M,k}$, $k = 1,..., n$, whereas $I^{(+)}$ is a matrix independent of z, which yields immediately the corresponding eigenvalues and eigenvectors, which are the only quantities of physical relevance for large times. These considerations also show that the non-Hermiticity of $I^{(+)}$ is a direct consequence of $\tilde{M}(z)$ depending on z and hence of the existence of lifetime effects.

In the above, we have assumed that $M(t)$ is Hermitian, which corresponds to assuming $\Omega = 0$ in Eq. (2.14). However, the general case may be treated in the same way by using a biorthogonal expansion already in Eq. (3.12).

3.4. Physical Interpretations. The Doorway Mode Concept

We have seen above that in typical physical situations $\tilde{C}(z)$ is analytic everywhere except for some singularities situated in the lower half-plane. Of these singularities, the n macroscopic poles typically are situated quite close to the real axis and are separated by a gap from the microscopic poles, which lie a distance of order t_c^{-1} further below in the lower half-plane of the physical sheet.

In the examples considered in the Appendix we find that the number of microscopic poles is equal to the number of macroscopic poles, i.e., n. This need not be the case in general. In fact, in order to satisfy higher order sum rules it often proves necessary to model $M(t)$ by a sum of exponentials or Gaussians or the like. Then, in general one may conclude that more microscopic than macroscopic poles will occur.

However, the detailed structure of the microscopic singularities is of no relevance to the results of the present paper. Of importance is only that for systems that can be modeled by memory kernels observing convenient decay properties one obtains macroscopic poles that are separated by a gap from the microscopic singularities. This gap reflects the existence of at least two different time scales and guarantees the existence of an autonomous

macrodynamics as expressed by Eqs. (2.23), (2.25), and (2.26). Moreover, the gap also allows the derivation of the generalized Green–Kubo relations given below.

It is customary[43] to associate the poles of $\tilde{C}(z)$ with modes. Obviously, then, the n macroscopic poles correspond to the long-lived collective modes of the system, the hydrodynamic modes, e.g., which are excited by the initial preparation of the system given by Eq. (2.3). Consequently, $C^{(+)}$ or $a^{(+)}$ describes just the decay of these modes, so that in close analogy to the point of view taken in quantum field theory, we may identify $z_{M,k}$, $k = 1,..., n$, with the complex energies and the pole strength Π_k with the corresponding normalization constants of the "wave functions" of these modes, Π_k (and hence $\Delta^{(+)}$) being different from 1 since the modes are not eigenstates of the Liouvillian of the system. This is an immediate consequence of the fact that L is a linear Hermitian operator or superoperator having real eigenvalues only, whereas the $z_{M,k}$ are complex. Thus, the modes may be viewed as a kind of pseudostate of L, the connection of which to real states such as $\rho(t_0)$ being established in terms of Π_k or $\Delta^{(+)}$.

In an isolated system the decay of these modes is due to their coupling with the extremely many irrelevant degrees of freedom which serve as a kind of reservoir. It has been suggested recently by Zeh[45] that the corresponding transfer of macroscopic information into the irrelevant channels should proceed by way of a two-step procedure similar to the doorway state mechanism in nuclear physics. This suggests that the microscopic poles be associated with *doorway modes*. These may be understood as short-lived collective excitations mediating the information transfer. This doorway-mode concept might be of some relevance in the foundation of so-called extended irreversible thermodynamics (EIT), which has found increasing interest recently (see, e.g., Ref. 46). In fact, assuming the number of microscopic poles is just n, then one easily convinces oneself that the contribution of the $2n$ poles may be taken into account exactly in an autonomous system of n differential equations of *second order* of the type considered in EIT, the so-called relaxation terms directly corresponding to the doorway modes, so that the Maxwell–Cattaneo[49] equations, e.g., are simply a consequence of the decay of the latter. Moreover, the fact that EIT predicts a finite velocity of the propagation of thermal disturbances is understood by noting that the response of the system to a macroscopic disturbance takes place via the excitation of doorway modes. The details of these considerations will be given elsewhere.

4. GENERALIZED GREEN–KUBO RELATIONS OF THE TIME-LOCAL PICTURE

One of the main aims of any theory of irreversible processes consists in giving explicit expressions for macroscopic quantities in microscopic terms. In the convolution picture (CP) the transport kernel $M(t)$ or $\tilde{M}(z)$ is considered as the macroscopic quantity of interest, since it may be interpreted as the matrix of generalized transport coefficients, which are time- or frequency-dependent, respectively. Equation (2.15), therefore, may be interpreted as a generalized Green–Kubo relation expressing M^{ir} as a correlation function of projected microscopic currents.

However, as discussed above, the decay of the long-lived modes is completely determined by the quantities $I^{(+)}$ and $\Delta^{(+)}$, so that in particular $I^{(+)}$ is the actual matrix of macroscopic transport coefficients. The aim now consists in finding convenient expressions relating $I^{(+)}$ and $\Delta^{(+)}$ to the correlation matrix.

4.1. Derivation of Generalized Green–Kubo Relations of the Time-Local Picture

Let us assume that there is a gap between macroscopic and microscopic singularities, i.e., there exist different time scales in the system. Then, the straight line $\bar{C}$ figuring in Eq. (3.1b) may be deformed in the way given by Fig. 2, so that we may write

$$C^{(+)}(t) = -\oint_{\mathrm{Ma}} \frac{dz}{2\pi} e^{-izt} \tilde{C}(z) \tag{4.1a}$$

and

$$C^{(m)}(t) = \int_{\mathrm{Mi}} \frac{dz}{2\pi} e^{-izt} \tilde{C}(z) \tag{4.1b}$$

where Ma is a closed contour encircling counterclockwise just the macroscopic singularities, whereas Mi is a (straight) line passing between the macroscopic and microscopic singularities. Now, for $t=0$ we obtain from Eqs. (4.1a) and (2.25)

$$\Delta^{(+)} = -\oint_{\mathrm{Ma}} \frac{dz}{2\pi} \tilde{C}(z) \tag{4.2}$$

Considering $\dot{C}^{(+)}(t)$, we obtain in the same way

$$\mathscr{L} := I^{(+)} \Delta^{(+)} = \oint_{\mathrm{Ma}} \frac{dz}{2\pi} z \tilde{C}(z) \tag{4.3}$$

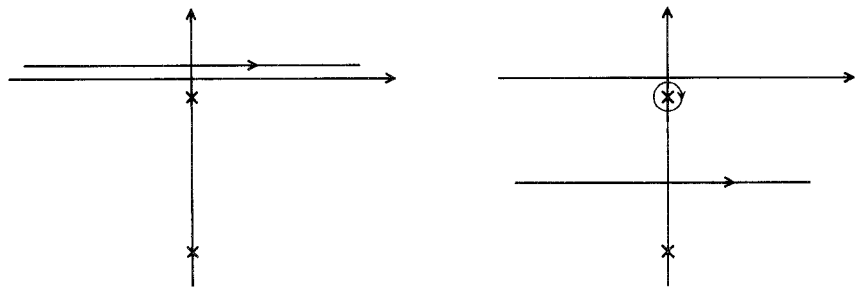

Fig. 2. The deformation of (a) the contour $\bar{C}$ into (b) the contours Ma (circle) and Mi (straight line). The positions of the microscopic and macroscopic poles are identified by the crosses. The case of a single observable is considered, the representation of the general case resting upon the spectrally resolved expression (3.12), i.e., the figure represents either one of the n terms occurring in Eq. (3.12). The derivations in the text are to be understood in this sense.

defining a new quantity $\mathscr{L}$, which will be shown in a further paper to play the role of the Onsager kinetic coefficient in the generalized irreversible thermodynamics to be developed there.

Equations (4.2) and (4.3) can be reformulated in such a way that a direct expression for the dissipative quantities $\mathscr{L}^{\mathrm{ir}}$ and $\Gamma^{(+)}$ can be obtained, where

$$\mathscr{L} = i\Omega + \mathscr{L}^{\mathrm{ir}}, \qquad \Delta^{(+)} = F + \Gamma^{(+)}, \qquad \Gamma^{(+)} = O(\xi) \tag{4.4}$$

Using $-\tilde{X}(z) = X(0) + iz\tilde{X}(z)$, we obtain first

$$\Delta^{(+)} = i \oint_{\mathrm{Ma}} \frac{dz}{2\pi} \frac{1}{z} [\tilde{C}(z) + C(0)] \tag{4.5}$$

If we now introduce a new contour $\overline{\mathrm{Ma}}$ chosen to encircle the macroscopic singularities *and* the point $z = 0$, we may rewrite this by means of Eq. (2.6),

$$\Gamma^{(+)} = -i \oint_{\overline{\mathrm{Ma}}} \frac{dz}{2\pi} \frac{1}{z} \tilde{C}(z) \tag{4.6a}$$

or, using

$$\oint_{\overline{\mathrm{Ma}}} dz\, z^{-2} = 0$$

we have

$$\Gamma^{(+)} = \oint_{\overline{\mathrm{Ma}}} \frac{dz}{2\pi} z^{-2} \tilde{\tilde{C}}(z) \tag{4.6b}$$

By similar manipulations we obtain from Eq. (4.3)

$$\mathscr{L}^{\mathrm{ir}} = i \oint_{\overline{\mathrm{Ma}}} \frac{dz}{2\pi} z^{-1} \tilde{\ddot{C}}(z) \tag{4.7}$$

The matrix of transport coefficients I^{ir}, where $I^{\mathrm{ir}} = I^{(+)} - i\Omega F^{-1}$,

$$I^{(+)} = \mathscr{L} \Delta^{(+)-1} = \sum_{n=0}^{\infty} \mathscr{L}(-F^{-1} I^{(+)})^n F^{-1} \tag{4.8a}$$

is obtained from Eqs. (4.6) and (4.7) as

$$I^{\mathrm{ir}} = \mathscr{L}^{\mathrm{ir}} F^{-1} - i \sum_{n=1}^{\infty} \oint_{\overline{\mathrm{Ma}}} \frac{dz}{2\pi} z^{-1} [\tilde{\ddot{C}}(z) - i\Omega] F^{-1} \times \left[\oint_{\overline{\mathrm{Ma}}} \frac{du}{2\pi} u^{-2} \tilde{\ddot{C}}(u) F^{-1} \right]^n \tag{4.8b}$$

the series being rapidly converging for small ξ, since $I^{(+)} = O(\xi)$ (see below).

Equations (4.6b)–(4.8) express all of the macroscopic quantities figuring in the time-local picture (TLP) in terms of $\tilde{\ddot{C}}(z)$, i.e., in terms of correlation functions of microscopic currents. We therefore shall call them *generalized Green–Kubo relations of the TLP.*

It is interesting to note that we may replace in Eq. (4.7) $\tilde{\ddot{C}}$ by its microscopic part $\tilde{\ddot{C}}^{(m)}$. In fact, using Eq. (3.14), we find

$$\tilde{\ddot{C}}^{(+)} = -i \sum_{k=1}^{n} \frac{z_{M,k}^2}{z - z_{M,k}} \Pi_k \tag{4.9}$$

so that by means of

$$\oint_{\overline{\mathrm{Ma}}} dz \, [z(z - z_{M,k})]^{-1} = 0$$

we conclude

$$\oint_{\overline{\mathrm{Ma}}} \frac{dz}{2\pi} z^{-1} \tilde{\ddot{C}}^{(+)}(z) = 0$$

and hence

$$\mathscr{L}^{\mathrm{ir}} = i \oint_{\overline{\mathrm{Ma}}} \frac{dz}{2\pi} z^{-1} \tilde{\ddot{C}}^{(m)}(z) = -\tilde{\ddot{C}}^{(m)}(0) = -\int_0^{\infty} dt \, \ddot{C}^{(m)}(t) \tag{4.10a}$$

since $\tilde{\ddot{C}} = \tilde{\ddot{C}}^{(+)} + \tilde{\ddot{C}}^{(m)}$ and $\tilde{\ddot{C}}^{(m)}(z)$ has no singularities inside $\overline{\mathrm{Ma}}$.

In an analogous way, we also obtain from Eq. (4.6b)

$$\begin{aligned} I^{(+)} &= \oint_{\overline{\mathrm{Ma}}} \frac{dz}{2\pi} z^{-2} \tilde{\ddot{C}}^{(m)}(z) \\ &= i \frac{d}{dz} \tilde{\ddot{C}}^{(m)}(z) \mid_{z=0} \\ &= -\int_0^\infty dt\, t \ddot{C}^{(m)}(t) \end{aligned} \tag{4.10b}$$

In view of Eqs. (4.10), (2.23), (2.25) one may say that the behavior of $C(t)$ is completely determined by $C^{(m)}(t)$, which is microscopically based. In CP the fact that $C(t)$ is given in terms of $M(t)$ has been widely used for getting approximate expressions for C by modeling M. The corresponding approach in TLP would consist in directly modeling $C^{(m)}$, fitting the parameters with the short-time behavior or corresponding frequency sum rules as usual. This is as general as in CP, but has the additional advantage that one obtains C directly and need not solve Eq. (2.14) or carry out the inverse Laplace transformation of Eq. (3.4).

4.2. Remarks on Generalized Green–Kubo Relations in CP and TLP

In order to discuss the difference between the generalized Green–Kubo relations (GKR) of CP and TLP, we note that although both relations express the macroscopic quantities in terms of the fast or irrelevant part of the microscopic motion, the way in which this is achieved is different. In the CP, the matrix of frequency-dependent transport kernels $\tilde{M}(z)$ is given by correlation functions of *projected* microscopic currents, i.e., the systematic or slow part is directly projected out of the microscopic motion.

In the TLP relations given above, $\mathscr{L}^{\mathrm{ir}}$, for example, is expressed directly in terms of $\tilde{\ddot{C}}$, i.e., in terms of correlation functions of the full microscopic currents obeying Hamiltonian dynamics, the slow part being taken away here by the contour integration.

The fact that the above GKR are directly formulated in terms of the correlation functions is clearly an advantage from the computational point of view, since the latter can be evaluated by standard methods such as cluster or diagram expansions. This is not the case for $\tilde{M}(z)$ because of the appearance of the projected propagator. Moreover, it should be mentioned as a further advantage that Eqs. (4.6) and (4.7) directly yield the "memory" renormalized quantities, i.e., the real decay constants, whereas these are obtained in the CP only from the solution of Eq. (2.29).

The differences between the GKR of CP and TLP vanish if we consider the particular case of very slow processes for which the lifetime effects disappear. As is well known (cf. Ref. 43, §5.4) in this case the generalized Green–Kubo relation reduces to the conventional one, which expresses the transport or kinetic coefficients directly in terms of time correlation functions. It will be instructive to rederive this result from the relations given above.

4.3. Conserved Quantities

Let us assume there is a slowness parameter q, so that we may write

$$\dot{A}_k = qJ_k(q), \qquad k = 1,..., n \tag{4.11}$$

where $\dot{A}_k = iLA_k$ and $J_k(0) \neq 0$. This means that A_k is a conserved quantity in the limit $q = 0$. Using Eq. (4.11), we write

$$\tilde{\tilde{C}}(z) = q^2 Q(q, z) \tag{4.12}$$

where $Q(q, z)$ is the analytical continuation into the lower half-plane of the matrix of current–current correlation functions defined by

$$\langle J(q)| \frac{i}{z - L} |J(q)^+ \rangle, \qquad \text{Im } z > 0 \tag{4.13}$$

Introducing (4.11) and (4.12) into (4.6b) immediately yields

$$\Gamma^{(+)} = O(q^2) \tag{4.14}$$

Analogously, we obtain from (4.7)

$$\mathscr{L}^{\text{ir}} = O(q^2)$$

Now, let us assume for the time being that the observables A_k are all even with respect to time reversal, so that $\Omega = 0$. Using Eq. (4.8), we find

$$I^{\text{ir}} = \mathscr{L}^{\text{ir}} F^{-1} + O(q^4) \tag{4.15}$$

and

$$I^{(+)} = I^{\text{ir}} = q^2 \bar{I} + O(q^4) \tag{4.16}$$

which defines $\bar{I}$. Now, using Eq. (2.33), we find

$$\tilde{\tilde{C}}^{(+)}(z) = q^4 \frac{\bar{I}^2}{z + iq^2 \bar{I}} \Delta^{(+)}$$

so that the contribution of the macroscopic poles of $\tilde{\tilde{C}}$ to the contour integral of Eq. (4.7) is seen to be of order q^4. This means that for q suf-

ficiently small but fixed, we may shrink the contour $\overline{\mathrm{Ma}}$ in Eq. (4.7) to a circle around $z=0$ which no longer contains the macroscopic poles. The error committed in this way obviously is just of order q^4. Thus, we obtain from Eqs. (4.7), (4.15), and (4.16) $I^{\mathrm{ir}} = q^2 Q(q,0) + O(q^4)$. Now we may let $q \to 0$ in Q, so that

$$\bar{I} = \lim_{q \to 0} Q(q,0) + O(q^2) \tag{4.17}$$

Note that $Q(q,z)$ is obtained from Eq. (4.13) by way of analytical continuation, so that, for *finite* q, $Q(q,z)$ is still analytic around $z=0$ and (4.17) is well defined. If, however, we want to express $\bar{I}$ in terms of the explicit expression given by Eq. (4.13), we may use that

$$Q(q,0) = \lim_{z \downarrow 0} \langle J(q) | \frac{i}{z-L} | J(q)^+ \rangle$$

so that finally we obtain

$$\bar{I} = \lim_{q \to 0} \lim_{z \downarrow 0} \langle J(q) | \frac{i}{z-L} | J(q)^+ \rangle + O(q^2) \tag{4.18}$$

which is the well-known expression defining the matrix of transport coefficients of conserved quantities in terms of time correlation functions.

The more general case where $\Omega \neq 0$ can be treated in a similar manner. However, since $\Omega = O(q)$, the macroscopic poles now also contribute terms, which are of order q^2 and q^3. These must be accounted for explicitly when the contour is shrunk in the sense explained above. As a result, one obtains

$$\bar{I} = \lim_{q \to 0} \lim_{z \downarrow 0} \langle J^d(q) | \frac{i}{z-L} | J^d(q)^+ \rangle \tag{4.19}$$

where J^d is as usual the dissipative part of the current, i.e., we have

$$qJ^d(q) = i(L - \Omega)A \tag{4.20}$$

instead of Eq. (4.11).

5. ROUTE TO MICROSCOPIC CALCULATIONS

The generalized Green–Kubo relations (GKR) derived in Eqs. (4.6)–(4.8) may be used for carrying out microscopic calculations of macroscopic quantities $\mathscr{L}$, $I^{(+)}$, $\Delta^{(+)}$ in starting directly from the explicit expression

$$\tilde{\tilde{C}}(z) = -\langle \dot{A} | \frac{i}{z-L} | \dot{A}^+ \rangle = -\langle A | \frac{iL^2}{z-L} | A^+ \rangle \tag{5.1}$$

valid for $\mathrm{Im}\, z > 0$. Standard methods such as perturbation, cluster, or diagram expansions as applied to Eq. (5.1) typically yield a series of the structure

$$\tilde{\tilde{C}}(z)\, F^{-1} = i\Omega + \sum_{m=0}^{\infty} \frac{\tilde{b}_m(z)}{z^m} \tag{5.2}$$

where the coefficient functions $\tilde{b}_m(z)$ are related to definite microscopic events as explained below and Ω and F [cf. Eqs. (2.6) and (2.11)] have been introduced for later convenience.

Let us consider for the sake of illustrating Eq. (5.2) the case of a gas of classical particles interacting via short-range forces. Quite naively, one might be tempted to expand Eq. (5.1), as

$$\tilde{\tilde{C}}(z) = -i \sum_{n=0}^{\infty} z^{-(n+1)} \langle \dot{A} | L^n | \dot{A}^+ \rangle$$

which, however, does not exist for any value of z, since from $L = L_0 + L_1$ and $[L_0, L_1] \neq 0$ we may conclude that $\langle \dot{A} | L^{n+1} | \dot{A}^+ \rangle = O(n!)$. Instead, one must use conventional many-body techniques for evaluating Eq. (5.1).

For instance, by applying the Green–Cohen cluster expansion technique to the N-particle propagator $(z - L)^{-1}$ in Eq. (5.1) we readily arrive at Eq. (5.2). Thus, $b_0(t)$ is easily identified with the contributions due to a single binary (or a genuine multiple) collision, so that $b_0(t)$ decays rapidly on a time scale given by the collision time t_c and $\tilde{b}_0(z)$ is analytic for $|z| \leqslant O(t_c^{-1})$ indeed.

Besides these basic events, there are also uncorrelated sequences of such events. Thus, the contribution of m binary collisions, e.g., is contained in $\tilde{b}_{m+1}(z)$ in (5.2), where $b_m(t)$ again decays on the time scale given by t_c. Of course, there are many more contributions to (5.2). Of these, the so-called ring events, i.e., correlated sequences of binary collisions, lead to long-time tail phenomena, which need special treatment, as briefly sketched in Section 5.3.

The occurrence of terms that behave as z^{-m}, $m > 0$, for $z \to 0$ is usually called the problem of secular divergences and hinders the use of expansion (5.2) in conventional GKR, which involve $z \to 0$. This problem usually is circumvented by introducing the memory function formalism, which corresponds to a summation of Eq. (5.2) after which $z \to 0$ can be carried out in most cases. However, the appearance of the projected propagator [cf. Eq. (2.13b)] in $M(t)$ or $\tilde{M}(z)$ hinders the application of straightforward many-body techniques, so that $\tilde{M}(z)$ is not as easily expressed in microscopic terms as $\tilde{\tilde{C}}(z)$ or $\tilde{b}_m(z)$.

The problem of secular divergences and hence the necessity to introduce M does not arise in the present theory since our GKR (4.6)–(4.8) involve mesoscopic values of z only, i.e., where $O(t_R) \leqslant |z|^{-1} \leqslant O(t_c)$. For these z, the series (5.2) may well converge, so that our GKR allow us to express $\Gamma^{(+)}$, $\mathscr{L}$, and $\Gamma^{(+)}$ directly in terms of the $\tilde{b}_m$ and hence to relate the former in a direct way to the microscopic dynamics of the system.

5.1. General Scheme for Microscopic Calculations

Let us assume for the time being that Eq. (5.2) is uniformly converging for some mesoscopic values of z, i.e., inside some annulus U where $r \leqslant |z| \leqslant R$, $r = O(t_R^{-1})$, and $R = O(t_c^{-1})$. Moreover, let us assume that $\tilde{b}_m(z)$ is analytic for $|z| \leqslant R$. Then[50] we may use Eq. (5.2) in our GKR (4.6)–(4.8) and evaluate the sums term by term. Thus, we obtain the desired relations as

$$\mathscr{L} = -\sum_{m=0}^{\infty} \frac{1}{m!} b_{m;m} F \tag{5.3}$$

$$\Gamma^{(+)} = i \sum_{m=0}^{\infty} \frac{1}{(m+1)!} b_{m;m+1} F \tag{5.4}$$

and

$$I^{(+)} = -\left(\sum_{m=0}^{\infty} \frac{1}{m!} b_{m;m} \right) \times \sum_{k=0}^{\infty} \left(-i \sum_{p=0}^{\infty} \frac{1}{(p+1)!} b_{p;p+1} \right)^k \tag{5.5}$$

where we have used the notation

$$f_{;n} = \lim_{z \to 0} \frac{d^n}{dz^n} f(z) = i^n \int_0^{\infty} dt\, t^n f(t) \tag{5.6}$$

so that $b_{m;n}$ are the Taylor coefficients of $\tilde{b}_m(z)$, i.e.,

$$\tilde{b}_m(z) = \sum_{n=0}^{\infty} \frac{1}{n!} b_{m;n} z^n \tag{5.7a}$$

Note that we have the order-of-magnitude estimate

$$b_{m;n} = O(R^{-n}); \qquad R = O(t_c^{-1}) \tag{5.7b}$$

due to a theorem of Cauchy.[51]

Equations (5.3)–(5.5) are the desired expressions relating the macroscopic parameters to the coefficient functions $\tilde{b}_m(z)$, the $b_{m;n}$ corresponding to moments of $b_m(t)$ accoding to Eq. (5.6).

For applications the following order-of-magnitude estimates are of utmost importance. Upon introducing Eqs. (5.7) into Eq. (5.2), we find, for $|z| \gtrsim r$,

$$\tilde{\tilde{C}} = \sum \tilde{b}_m(z)\, z^{-m} = \sum b_{m;0} z^{-m}[1 + O(r/R)]$$

This is a Taylor series in terms of $u = z^{-1}$. Hence, we may apply Cauchy's theorem of Taylor coefficients[51] again to obtain

$$b_{m;0} = b_{0;0} O(t_R^{-m}) \tag{5.8}$$

Using $\mathscr{L} = O(t_R^{-1})$ and (5.7b), we find, observing $r = O(t_R^{-1})$ and $R = O(t_c^{-1})$,

$$b_{m;n} = O(t_c^n / t_R^{m+1}) \tag{5.9}$$

This relation is to be understood in the sense that it yields the asymptotic behavior of $b_{m;n}$ for $t_c \to 0$ and $t_R \to \infty$. In this sense, Eqs. (5.3) and (5.4) obviously represent expansions in ascending order of $\xi = t_c/t_R$ and Eq. (5.5) can be rearranged into such an expansion, which we call a *time scale expansion* (TSE) in the following. TSEs have proved useful recently in several branches of statistical physics, since ξ is a universal parameter, which is very small for many processes of interest, so that TSEs usually are rapidly converging.

If microscopic and macroscopic time scales are sufficiently well separated so that terms of order ξ, i.e., lifetime effects, may be neglected altogether, one obtains the very simple result

$$\mathscr{L} F^{-1} = I^{(+)} = -b_{0;0} \tag{5.10}$$

Neglecting lifetime effects corresponds to taking the so-called Markovian limit in the CP approach. Hence, if one is allowed or willing to work in this limit (as is very often done in practice), one obtains the very simple result that in Eq. (5.2) one simply has to drop all terms that are diverging for $z \to 0$ and consider the remainder at $z = 0$.

Correction terms over this rule are easily obtained from Eqs. (5.3)–(5.5), for instance,

$$\mathscr{L} F^{-1} = -b_{0;0} - b_{1;1} + O(\xi^2) \tag{5.11a}$$

$$I^{(+)} F^{-1} = i b_{0;1} + O(\xi^2) \tag{5.11b}$$

$$I^{(+)} = -b_{0;0} - b_{1;1} + i b_{0;0} b_{0;1} + O(\xi^2) \tag{5.11c}$$

where lifetime effects are included in leading order here.

An important special case of Eq. (5.2) arises if the coefficient functions obey the factorization property

$$i\tilde{b}_m(z) = [i\tilde{b}_0(z)]^{m+1} \tag{5.12}$$

Then, we obviously may rewrite the above expressions in terms of the Taylor coefficients $b_{0;m}$ of $\tilde{b}_0(z)$ alone. For example, we obtain instead of Eq. (5.11c)

$$I^{(+)} = -b_{0;0} - ib_{0;1}b_{0;0} + O(\xi^2) \tag{5.13}$$

since $b_{1;1} = i(b_{0;1}b_{0;0} + b_{0;0}b_{0;1})$. Higher order terms of the TSE are obtained from Eqs. (5.6) and (5.12) by grouping together all terms with an equal number of factors. The above results also allow one to establish explicitly the connection between CP and TLP quantities. In fact, if (5.12) holds, Eq. (5.2) may easily be resummed to yield

$$\tilde{\tilde{C}}F^{-1} = i\Omega + \tilde{b}_0 + i\tilde{b}_0^2(z - i\tilde{b}_0)^{-1}$$

so that, from comparing with Eq. (3.4), we obtain

$$\tilde{b}_0(z) = -\tilde{M}(z) \tag{5.14}$$

As a consequence of Eq. (5.14) we may use Eqs. (5.3), (5.4), and (5.6) directly for expressing $\mathscr{L}$, $\Gamma^{(+)}$, and $I^{(+)}$ in terms of $\tilde{M}(z)$. In particular, Eq. (5.6) yields in this way the solution of Eq. (2.29). This has been obtained by several authors,[30,47,52] and our result can be shown to agree with the known results term by term.

The expressions (5.3)–(5.5) and (5.11) are valid in full mathematical rigor provided only Eq. (5.2) is uniformly converging inside some annulus $r \leqslant |z| \leqslant R$ and that Eq. (5.7a) exists for $|z| \leqslant R$. Moreover, our set of observables A has to be a complete one, i.e., the number of macroscopic poles agrees with the number of observables. This is a general requirement both in CP and TLP. A criterion for checking this point will be given below Eq. (5.20).

5.2. Laurent Series Expansion of Correlation Matrix

Additional insight into expansions of the kind (5.2) can be gained by introducing the Taylor expansion (5.7a) of $\tilde{b}_m(z)$ into Eq. (5.2). By conveniently rearranging the resulting expression, one obtains a series in positive and negative powers of z,

$$\tilde{\tilde{C}}(z) = \sum_{n=-\infty}^{\infty} \zeta_n z^n \tag{5.15a}$$

Using

$$\tilde{\tilde{C}}(z) = -i\Omega F + izF - z^2\tilde{C}(z)$$

one obtains correspondingly

$$\tilde{C}(z) = \sum_{n=-\infty}^{\infty} \alpha_n z^n \tag{5.15b}$$

Equations (5.15) obviously are Laurent expansions, which represent $\tilde{\tilde{C}}$ and $\tilde{C}$ inside the annulus U passing between the macroscopic and microscopic singularities (cf. Fig. 3).

From the theory of complex functions we recall that Laurent's expansion inside its annulus of convergence is unique, uniformly converging, and may be differentiated and integrated term by term. The coefficients are obtained according to the standard rule,

$$\zeta_n = \oint_{C'} \frac{dz}{2\pi i} z^{-(n+1)} \tilde{\tilde{C}}(z) \tag{5.16}$$

where C' denotes a closed contour, which lies entirely in the annulus and encircles the origin counterclockwise.

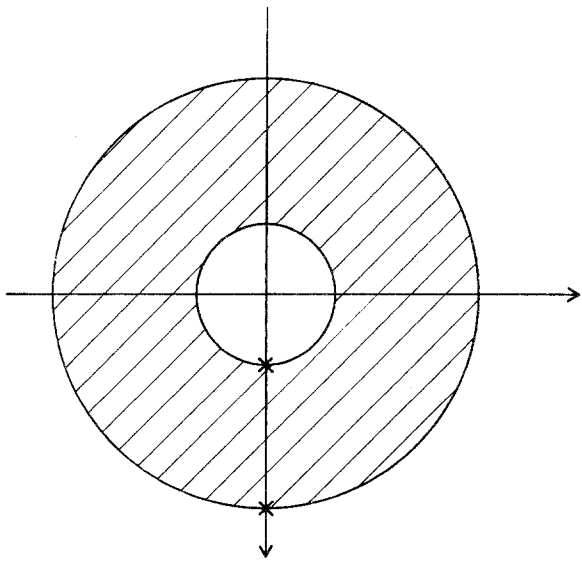

Fig. 3. The annulus U (shaded area) as introduced in the text. In the case of several observables the figure is to be understood as explained with regard to Fig. 2.

The existence of the Laurent expansions (5.15) is hence guaranteed if there is a gap between the macroscopic and microscopic singularities and vice versa. If using the CP approach, this means that Eqs. (5.15) exist if $M(t)$ has the decay properties proposed in Section 3. Laurent's expansion may be split into its regular and principal parts. Accordingly, we may write, e.g.,

$$\tilde{C}(z) = \sum_{n=-1}^{-\infty} \alpha_n z^n + \sum_{n=0}^{\infty} \alpha_n z^n =: \tilde{C}^{(+)} + \tilde{C}^{(m)} \tag{5.17}$$

so that the principal (regular) part corresponds to the macroscopic (microscopic) part of the correlation matrix [cf. Eq. (2.23)]. Equation (5.17) may be obtained by using Eq. (5.16),

$$\tilde{C}^{(+)} = \frac{i}{z + iI^{(+)}} \Delta^{(+)} \tag{5.18}$$

and the fact that $\tilde{C}^{(m)}$ has no macroscopic singularities. According to Eqs. (5.17) and (5.18), the macroscopic information is fully contained in the principal part of the Laurent expansion of $\tilde{C}(z)$. On the other hand, we find from our GKR (4.6)–(4.8) and (5.15a)

$$\mathscr{L}^{\mathrm{ir}} = -\zeta_0, \qquad I^{(+)} = i\zeta_1 \tag{5.19}$$

so that the complete macroscopic information is contained in the coefficients ζ_0, ζ_1 of the regular, i.e., microscopic part $\tilde{\tilde{C}}^{(m)}$ of $\tilde{\tilde{C}}$. Note that Eq. (5.19) is in complete agreement with Eq. (4.10). Exploiting the properties of the Laurent expansion, we may now formulate a criterion of reliability of the approach to microscopic calculations as given in Section 5.1. By means of Eqs. (5.15a), (5.16) and (5.18) we easily derive for $n \geqslant 1$

$$\oint \frac{dz}{2\pi i} z^{(n-1)} \tilde{\tilde{C}}(z) = -(-iI^{(+)})^n \mathscr{L} \tag{5.20}$$

Then, using Eqs. (5.3)–(5.5) and introducing Eq. (5.2) into Eq. (5.20), we may derive criteria that the $\tilde{b}_m$ must obey. This procedure is greatly simplified by using Eq. (5.9) to obtain corresponding TSEs. In lowest order, we thus derive in the limit of small ξ

$$ib_{m;0} = (ib_{0;0})^m + O(\xi) \tag{5.21}$$

which agrees with Eq. (5.12) taken at $z = 0$.

5.3. Long-Time Tail Phenomena

In practical applications the set A in many cases is not a complete one. In fact, given a primitive set of slow variables $A_1,..., A_n$ one can always combine these into so-called multilinear variables, which are also slow. If these are explicitly included into A, the present theory is applicable. If not, one observes an additional spectrum of modes. In an infinitely extended system this spectrum is a continuous, one so that, besides the macroscopic poles, we also observe branch cuts in $\tilde{M}(z)$ or $\tilde{C}(z)$ corresponding, e.g., to a $z^{1/2}$ behavior in a three-dimensional fluid. These branch cuts typically begin at $z=0$ or at a hydrodynamic frequency and may extend to $z=\infty$. Hence, our theory is not applicable immediately, since there is no annulus U inside which $\tilde{C}$ is analytic. However, many-valued functions are studied best on the complete Riemannian surface, which, for $z^{1/2}$, say, consists of two sheets with a branch *point* at $z=0$ corresponding to an algebraic singularity.

If represented in this way, $\tilde{C}(z)$ displays both poles and algebraic singularities as macroscopic singularities, which again are separated by a gap from the microscopic singularities. Accordingly, instead of Eq. (2.23), $\tilde{C}(z)$ may now be split into *three* different terms corresponding to the decay of transients (doorway modes, e.g.), the decay of macroscopic modes governed by the autonomous macrodynamics, and the long-time tail phenomena contributed by the algebraic singularities.

The gap between macroscopic and microscopic singularities allows us to modify most of the results of the present paper so as to include long-time tail phenomena. In fact, Laurent's expansion and Eq. (5.2) are easily extended to cover the case of algebraic singularities by allowing for fractional powers of z. From this, as will be shown in a subsequent paper,[53] a scheme for carrying out microscopic calculations may be developed much in the same way as in Section 5.1.

It is instructive to discuss the relation between CP and TLP approaches in this context. We note that with long-time tails occurring, $\tilde{M}(z)$ also has an algebraic (i.e., macroscopic) singularity. Thus, $\tilde{M}(z)$ is not purely microscopically based. Moreover, Eqs. (2.28), and (2.29) have no solution, whereas the corresponding solutions of the dispersion relations [cf. Eq. (3.13)] and hence $I^{(+)}$ well exist. Then, by applying the theory of residua to Eq. (3.4) one arrives at the splitting of $C(t)$ into the three different terms mentioned above. These are mixed up in the CP equations of motion (2.14), and hence in the notion of the frequency-dependent transport coefficients, in an intricate way, whereas we prefer to restrict the notion of transport to the autonomous macrodynamics corresponding to the decay of macroscopic modes.

6. GENERALIZED GREEN–KUBO RELATIONS IN TIME REPRESENTATION

In Sections 4 and 5 we have derived generalized Green–Kubo relations (GKR) and corresponding time scale expansions (TSE) which directly relate transport quantities to $\tilde{\tilde{C}}(z)$. However, these formulas are not immediately applicable if the correlation functions are given in time representation as a result of a measurement, for example. Therefore, we shall now derive a TSE for $I^{(+)}$, $\mathscr{L}$, and $\Delta^{(+)}$ in time representation. This also will provide us with a way for relating $I(t)$ [cf. Eq. (2.10)] to the short-time behavior of the time correlation functions.

Let us consider the case again that $\Omega = i\dot{C}(0) = 0$ and use Eq. (2.10), i.e. $(t_0 = 0)$

$$I(t) = I^{\mathrm{tr}}(t) = \dot{C}(t)\, C^{-1}(t) \tag{6.1}$$

Now we write

$$\dot{C}(t) = \int_0^t dt'\, \ddot{C}(t') \tag{6.2}$$

and, using Eq. (2.6),

$$C(t) = F + \int_0^t dt'\, (t - t')\, \ddot{C}(t') =: F + g(t) \tag{6.3}$$

which is easily proved by partial integration. On the other hand, we obtain from using Eqs. (2.23), (2.25), and (4.10) in Eq. (6.3) that

$$g(t) = O(t/t_R) \tag{6.4}$$

which is valid for $0 < t \lesssim t_R$.

Thus, for not too large t we may write

$$I(t) = \sum_{n=0}^{\infty} I_n(t)$$

where

$$I_n(t) = (-)^{n+1} \int_0^t dt'\, \ddot{C}(t')\, F^{-1} \left[\int_0^t dt''\, (t - t'')\, \ddot{C}(t'')\, F^{-1} \right]^n \tag{6.5}$$

which by virtue of Eq. (6.4) is already the desired TSE. In order to find the asymptotic value $I^{(+)}$ [cf. (2.17)], we assume that the time scales given by

t_c and t_R are sufficiently well separated that we may find a time t^0, say, that may be of the order of several t_c, so that

$$I(t^0) = I^{(+)} = \sum_{n=0}^{\infty} I_n, \qquad I_n := I_n(t^0) \tag{6.6}$$

with any required accuracy. Then, we obtain from Eq. (6.5), for example,

$$\begin{aligned} I_0 &= -\int_0^{t^0} dt'\, \ddot{C}(t')\, F^{-1} \\ I_1 &= +\int_0^{t^0} dt' \int_0^{t^0} dt''\, (t^0 - t'')\, \ddot{C}(t')\, F^{-1} \ddot{C}(t'')\, F^{-1} \\ &\vdots \end{aligned} \tag{6.7}$$

For obtaining the related TSE for $\Gamma(t)$, where

$$\Delta(t) = F + \Gamma(t) := e^{I^{(+)}t} C(t)$$

so that $\Gamma^{(+)} = \lim_{t\to\infty} \Gamma(t)$, we use Eq. (6.3), from which

$$\Gamma(t) = g(t) + \sum_{n=1}^{\infty} \frac{(I^{(+)}t)^n}{n!} [1 + g(t)] \tag{6.8a}$$

Now, introducing $I^{(+)}$ of Eq. (6.6) and collecting the terms that contain an equal number of correlation functions C yields the TSE for $\Gamma(t)$.

For example, we obtain for the asymptotic value $\Gamma^{(+)}$ in lowest order

$$\Gamma^{(+)} = \Gamma(t^0) = -\int_0^{t^0} dt'\, t' \ddot{C}(t') = O(\xi) \tag{6.8b}$$

For $\mathscr{L} = I^{(+)} \Delta^{(+)}$ we obtain consequently

$$\mathscr{L} = -\int_0^{t^0} dt'\, \ddot{C}(t') \left[1 - t^0 F^{-1} \int_0^{t^0} dt''\, \ddot{C}(t'') + O(\xi^2) \right] \tag{6.9}$$

where lifetime effects are included explicitly in order of ξ. The above formulas relate the asymptotic values of pertinent macroscopic quantities to the short-time behavior of the time correlation functions $\ddot{C}(t)$, which may also be expressed in terms of current–current correlation functions. The expansions are obtained as TSE, where, for example,

$$I_{n+1} = I_n [1 + O(\xi) + O(t^0/t_R)] \tag{6.10}$$

Thus, if we include only a finite number of terms, we may not let $t^0 \to \infty$ in these series. Instead, we have to choose t^0 sufficiently large so that (6.6) is

valid and consider a sufficient number of terms so that convergence is achieved, the number of terms necessary increasing for increasing t^0 because of Eq. (6.10).

The above TSE are valid for the special case $\Omega = 0$, i.e., that all observables A_k, $k = 1,..., n$, are even with respect to time reversal. The more general case may be treated essentially in the same way if we repeat the above derivations in a kind of interaction representation obtained by replacing $C(t)$ with $C_W(t) = e^{i\Omega t}C(t)$ and using

$$\exp(i\Omega t)\exp(-I^{(+)}t) = \exp_{(-)}\left[\int_0^t dt'\, I_W(t')\right]$$

where $I_W(t) = e^{i\Omega t}I^{ir}e^{-i\Omega t}$. The details will be given elsewhere.

Applications of the above expressions may be found in Ref. 40, where Eq. (6.7) is evaluated in the special case of kinetic theory. It is shown there that the role of I_1 consists in cancelling the secularly divergent contribution $\sim t$ contained in I_0 and to produce the correction term to the Boltzmann equation, which arises from the finite duration of a binary collision and was found earlier by several authors (cf. Ref. 48).

7. SUMMARY AND CONCLUSIONS

In the present paper, we considered systems for which two (or several) different time scales exist, corresponding to "slow" (t_R) and "rapid" (t_c) degrees of freedom. In the convolution picture (CP) this implies the decay property (2.18) of the memory kernel $M(t)$ provided set A is complete in the space of slow variables, which we assume throughout. This might seem a severe restriction for applications in fluid systems where long-time tail phenomena are observed. However, this restriction is overcome, as briefly discussed in Section 5.3.

The aim of the paper was to develop the time-local picture (TLP) of systems with different time scales, taking full account of lifetime (memory, non-Markovian) effects resulting from incomplete separation of time scales. The basic observation for the TLP philosophy is comprised in the additive splitting $C(t) = C^{(+)}(t) + C^{(m)}(t)$ [cf. Eq. (2.23)], the consistency of which with CP is given in Section 3. In Eq. (2.23), $C^{(m)}$ represents the initial slip of the correlation matrix $C(t)$ resulting from the decay of transients, the doorway modes, e.g. Hence, the decay of $C(t)$ or of $a(t)$ [cf. Eq. (2.27)] is given at macroscopic times $t \gg t_c$ by the macroscopic branch $C^{(+)}$ of C. The time evolution of $C^{(+)}$ is completely specified by $I^{(+)}$ and $\Delta^{(+)}$ [cf. Eq. (2.25)], which are the only quantities of macroscopic interest, $I^{(+)}$ representing the matrix of transport or decay coefficients, whereas $\Delta^{(+)}$

establishes the relation between the autonomous macrodynamics obeyed by $C^{(+)}$ and the initial preparation of the system.

We have presented a new set of generalized Green–Kubo relations (GKR) in Section 4, which are rigorous expressions relating $I^{(+)}$ and $\Delta^{(+)}$ directly to the physical correlation functions obeying Hamiltonian dynamics. A typical example for the application of our GKR has been given in Section 5 in applying it to so-called secularly divergent expressions of $\tilde{C}(z)$. These may arise from analyzing $\tilde{C}(z)$ in terms of sequences of binary collisions, in which case our GKR lead in a straightforward way to a time scale expansion relating $I^{(+)}$ and $\Delta^{(+)}$ to the properties of such binary collisions, including lifetime effects, in-medium corrections, e.g.

The TLP approach developed concentrates on the macroscopic decay properties given in terms of $I^{(+)}$, $\Delta^{(+)}$, or $\mathscr{L}$. Of course, these quantities also can be obtained from CP [cf. Eq. (2.29)], so that the two approaches are equivalent in this sense. However, as shown by way of examples,[22–29] they may give very different results in approximations. It would be interesting to test this point by using our GKR, since these directly generate expansions (perturbation, density, or the like) for $I^{(+)}$ from corresponding expansions of the correlation matrix.

APPENDIX. SOLUTIONS OF THE DISPERSION RELATION $iz = \tilde{M}(z)$

Let us consider the case that our set $A = \{A_1, ..., A_n\}$ consists of a single variable only, so that $M(t)$ is a real, scalar function. We first treat the case that $M(t)$ is given by an exponential, i.e., we put

$$M(t) = (1/t_c t_R)\, e^{-t/t_c} \tag{A1}$$

t_c and t_R having been defined in Eqs. (2.17). From (A1) we obtain

$$\tilde{M}(z) = i t_R^{-1}/(z t_c + i) \tag{A2}$$

which may be considered as the representation of $\tilde{M}(z)$ on the full physical sheet.

The solutions of the dispersion relation (3.5), i.e.,

$$iz = \tilde{M}(z) \tag{A3}$$

are given by the solutions z_M, z_m of

$$z^2 + \frac{i}{t_c} z - \frac{1}{t_c t_R} = 0 \tag{A4}$$

yielding

$$z_M = -\frac{i}{2t_c}[1-(1-4\xi)^{1/2}] = -\frac{i}{t_R}(1+\xi+2\xi^2+\cdots) \tag{A5}$$

and

$$z_m = -\frac{i}{2t_c}[1+(1-4\xi)^{1/2}] = -\frac{i}{t_c}(1-\xi-\xi^2-\cdots) \tag{A6}$$

Thus, the actual decay or transport coefficient is

$$I^{(+)} = \frac{1}{2t_c}[1-(1-4\xi)^{1/2}] = \frac{1}{t_R}(1+\xi+2\xi^2+\cdots) \tag{A7}$$

For the present case, we may also calculate explicitly the residue of $\tilde{C}(z)$, which after some manipulations may be written as

$$\mathrm{Res}(z_M) = i\frac{z_m}{z_m - z_M} = i(1+\xi+3\xi^2+\cdots) \tag{A8}$$

and

$$\mathrm{Res}(z_m) = i\frac{z_M}{z_M - z_m} = i(\xi+3\xi^2+\cdots) \tag{A9}$$

Using (A8) together with Eq. (3.8), we obtain therefore

$$I^{(+)} = -i\,\mathrm{Res}(z_m) = \xi+3\xi^2+\cdots \tag{A10}$$

which may also be obtained by using Eq. (A2) in Eq. (5.4). A physical interpretation of Eq. (A10) is obtained by noting that $\mathrm{Res}(z_m)$ just corresponds to the strength of the doorway mode. In the example considered, the doorway mode is the only transient, since there is no further singularity even at $z=\infty$. Thus, the doorway mode alone is responsible for the initial slip, which explains Eq. (A10).

As a further example, we assume $M(t)$ to be given by the Gaussian

$$M(t) = (2/\pi^{1/2}t_c t_R)\exp[-(t/t_c)^2] \tag{A11}$$

Introducing $b = 2(\pi^{1/2}t_R)^{-1}$ and $2v = izt_c$, we obtain from Eq. (A3) for $M[v] := \tilde{M}(z)$

$$M[v] = b\int_0^\infty dt\,\exp(2vt-t^2) \tag{A12}$$

Putting $v = v' + iv''$, we write the integral as

$$\int_0^\infty dt \exp(2v't - t^2 + 2iv''t)$$

$$= [\exp(v'^2)] \int_0^\infty dt \exp[-(t-v')^2 + 2iv''t]$$

$$= [\exp(v'^2)] \int_{-v'}^\infty dt\, [\exp(-t^2)]\{\cos[2v''(t+v')]$$

$$+ i \sin[2v''(t+v')]\} \tag{A13}$$

We are now going to show first that the dispersion relation (A13) or

$$v = \tfrac{1}{2} t_c \tilde{M}[v] \tag{A14}$$

can have solutions only on the imaginary axis of the complex z plane, which corresponds to $v'' = 0$. In order to show this, we assume v is a solution of (A14) with $v'' \neq 0$. Using (A13) we may obtain from (A14) the two equations valid for $v'' \neq 0$

$$v'v'' = (\xi/\sqrt{\pi})[\exp(v'^2)] \int_{-v'v''}^\infty dt$$

$$\times \{\exp[-(t/v'')^2]\} \cos[2(t + v'v'')] \tag{A15a}$$

$$v''^2 = (\xi/\sqrt{\pi})[\exp(v'^2)] \int_{-v'v''}^\infty dt$$

$$\times \{\exp[-(t/v'')^2]\} \sin[2(t + v'v'')] \tag{A15b}$$

where $\xi = t_c/t_R$.

Using $\tilde{M}^*(z) = \tilde{M}(-z^*)$, since $M(t)$ is real, we find that if z is a solution of the dispersion relation, so is $-z^*$, i.e., the solutions, if they exist at all, must lie symmetrically to the imaginary axis. This means that if v', v'' is a solution of Eqs. (A15), so is v', $-v''$. Using this result in Eqs. (A15) leads to two new equations

$$0 = \int_{-\infty}^\infty dt\, \{\exp[-(t/v'')^2]\} \cos[2(t + v'v'')] \tag{A16a}$$

$$0 = \int_{-\infty}^\infty dt\, \{\exp[-(t/v'')^2]\} \sin[2(t + v'v'')] \tag{A16b}$$

which must be fulfilled simultaneously if $v = v' + iv''$ is to be a solution of Eq. (A14). It is easy to show that (A16a) is fulfilled only if $2v'v'' = \pm k\pi$,

$k=0, 1, 2,...$, whereas from Eq. (A16b) we obtain the condition $2v'v''=\pm[2k+1)/2]\pi$. Since these two conditions contradict each other, we are led to the conclusion that there are no solutions with $v''\neq 0$. Thus, the only possible solutions correspond to $z=iz''$ or $v=v'$, $v'>0$, where, from Eqs. (A12) and (A13), we obtain the equation to be satisfied by v':

$$v'=\frac{\xi}{\sqrt{\pi}}[\exp(v'^2)]\int_{-v'}^{\infty} dt\exp(-t^2) \tag{A17}$$

or

$$\xi=2\frac{v'\exp(-v'^2)}{1+\mathrm{erf}(v')}=:U(v') \tag{A18}$$

where $\mathrm{erf}(\infty)=1$.

The graph of the function U defined by Eq. (A18) is given in Fig. 4, from which we may discuss the qualitative behavior of the solutions of the dispersion relation (A14). We note that for $\xi>\xi_0=0.522$, Eq. (A18) has no solution at all. As a consequence, the only singularity of $\tilde{C}(z)$ is the essential one at $z=\infty$. Thus, there is no exponentially decaying distribution to $C(t)$ and hence the autonomous equation (2.19) does not exist. For $\xi<\xi_0$ we find that Eq. (A18) has exactly *two* solutions v'_1, v'_2. For the case of very small ξ (very good separation of time scales) we find from Eq. (A18) the asymptotic expressions $v'_M=\xi/2$ and $v'_m=[\ln(1/\xi)]^{1/2}$. Thus, in this approximation the two poles of $\tilde{C}(z)$ are situated at

$$z_M=-i/t_R, \qquad z_m=i(2/t_c)[\ln(1/\xi)]^{1/2}$$

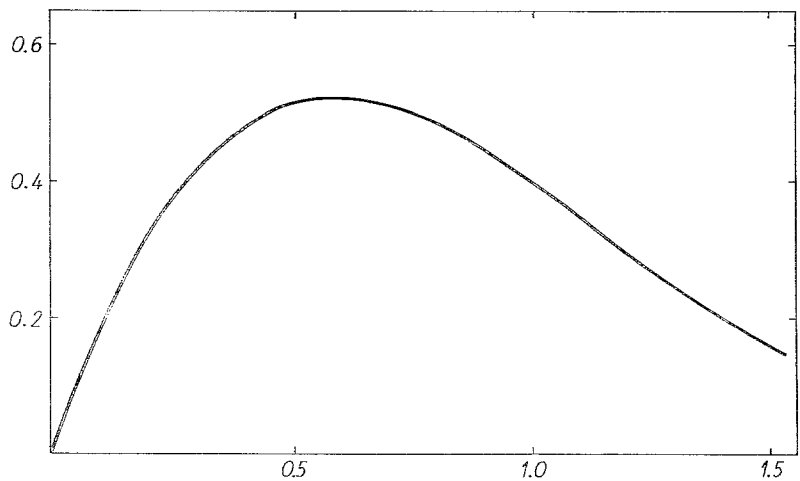

Fig. 4. The function $U(v)$ as introduced in Eq. (A18) versus v.

so that

$$z_m/z_M = (2/\xi)[\ln(1/\xi)]^{1/2}$$

these expressions being correct if ξ is sufficiently small, so that terms of order ξ and higher may be neglected.

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