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Moment and Continued Fraction Expansions of Time Autocorrelation Functions

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Two representations of the Laplace transforms of time autocorrelation functions, namely the moment expansion and Mori's continued fraction representation, are studied from the point of view of their convergence, with the aim of obtaining new general properties of time autocorrelation functions. First, the relation between them is established by using mathematical techniques in the case of a general dynamical variable, and a direct method, useful for applications, in the case of Hermitian variables. The mathematical structure associated to Mori's generalized random forces is investigated and it is shown that these random forces can be obtained by a Schmidt orthogonalization of the sequence of initial time derivatives of the dynamical variable considered. Then, the convergence criteria for both representations are examined and an illustration is given with the exactly solvable case of an isotopic impurity in a linear chain of coupled harmonic oscillators. Finally, the question of knowing whether the continued fraction expansion is convergent for any Hermitian dynamical variable and any system is discussed, with its implications for the general behaviour of time autocorrelation functions.

§ 1. Introduction

In two recent papers,[1),2)] Mori has developed a generalized Brownian motion theory of irreversible processes and then, with the purpose of studying the anomalous behaviour of damping constants and transport coefficients near second-order phase transition points, he was led to derive a continued fraction expansion for the Laplace transforms of time autocorrelation functions.

In order to explain the aims of the present work, let us first recall from Mori's formalism, the equations which will be our starting point.

We consider a dynamical variable $A(t)$, the invariant part of which is set to be zero, that is,

$$\lim_{T\to\infty}\frac{1}{T}\int_0^T A(t)\,dt=0\,. \tag{1·1}$$

We assume furthermore that $A(t)$ belongs to the Hilbert space of dynamical variables, the invariant parts of which are set to be zero and for which the inner product of the dynamical variable A with the Hermitian conjugate of the

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dynamical variable B is (A, B). For the largest part of this paper, we need not specify the form of the inner product; in addition to the usual properties $(A, B)=(B, A)^*$, $(A, A)\geq 0$, we shall only require for this scalar product to have the Liouville operator L Hermitian:

$$(A, LB)=(LA, B). \tag{1·2}$$

The starting point of Mori is to separate the time derivative $\dot{A}(t)=dA(t)/dt$ into a functional $F_1[A(s),\ t\geq s\geq$ initial time $t_0]$ depending upon the past history of $A(t)$, and an additional term $F_2(t, t_0)$ depending explicitly upon the other degrees of freedom; then, expanding the functional F_1 in terms of $A(s)$ and extracting the linear term, Mori defines the first random force $f_1(t)$ as the sum of $F_2(t, t_0)$ and the non-linear terms. Now the same procedure can be applied to $f_1(t)$ to define a second random force $f_2(t)$, then to $f_2(t)$ to define a third random force $f_3(t)$ and so on. In this way a hierarchy of random forces $f_j(t)$ is generated, the values of which at initial time $t_0=0$, which we denote by f_j, obey the recurrence equations

$$f_j=(1-\sum_{l=0}^{j-1}\mathcal{P}_l)iLf_{j-1}, \qquad f_0\equiv[A(t)]_{t=0}\equiv A, \tag{1·3}$$

where $\mathcal{P}_l$ is the projection operator onto the vector f_l. These vectors f_j form an orthogonal set,

$$(f_j, f_k)=0 \qquad j\neq k, \tag{1·4}$$

and evolve in time according to the equations

$$f_j(t)=\exp(iL_jt)iL_jf_{j-1} \qquad (j\geq 1), \qquad f_0(t)=A(t), \tag{1·5}$$

where

$$L_j=(1-\mathcal{P}_{j-1})L_{j-1}, \qquad L_0=L. \tag{1·6}$$

The initial time derivatives $\dot{f}_j=[df_j(t)/dt]_{t=0}$ satisfy the recurrence equations

$$\dot{f}_j=(1-\sum_{l=0}^{j-1}\mathcal{P}_l)iLf_j. \tag{1·7}$$

Denoting by $i\omega_j$ the projection of $\dot{f}_j$ onto the f_j axis,

$$i\omega_j=(\dot{f}_j, f_j)/(f_j, f_j), \tag{1·8}$$

we have

$$\dot{f}_j=i\omega_jf_j+f_{j+1} \tag{1·9}$$

and introducing the quantities

$$\Delta_j^2=(f_j, f_j)/(f_{j-1}, f_{j-1}) \tag{1·10}$$

the Laplace transform of the relaxation function $\Xi(t)=(A(t), A)/(A, A)$ can be written

$$\Xi(z)=\int_0^\infty (A(t),A)(A,A)^{-1}e^{-zt}dt$$
$$=\cfrac{1}{z-i\omega_0+\cfrac{\Delta_1^2}{z-i\omega_1+\cfrac{\Delta_2^2}{z-i\omega_2+\cdots}}}\ . \tag{1·11}$$

This expansion is Mori's infinite *continued fraction representation* for the Laplace transforms of time autocorrelation functions.

The knowledge of the analytic properties of the function represented by (1·11) is important from at least two points of view. First, the knowledge of the singularities is essential, since they determine the relaxation of $A(t)$. If one is going to describe the approach of $A(t)$ toward an equilibrium value, these singularities are expected to be located in the half-plane Re $z<0$. However, one should not think that the study of the analytic properties in the other half-plane Re $z>0$ lacks interest: indeed, as was also shown by Mori,[1] the expressions of the transport coefficients are proportional to the limiting value of $\Xi(z)$ when $z\to 0_+$. Therefore the knowledge of the convergence of the continued fraction has a practical interest, not to mention the theoretical interest in itself; indeed, as the theoretical interest is concerned, such a convergence study is in the line of recent efforts to study the convergence of equilibrium expansions such as the Virial expansion[3] or the analyticity of non-equilibrium expansions such that the density expansion of transport coefficients.[4]

Thus, one of the main purposes of the present paper is to study the convergence of two analytic representations of the Laplace transforms of time autocorrelation functions, namely the continued fraction (1·11) and the well-known *moment expansion*,[5] which is a power series expansion. Let us now also briefly recall the features of the moment expansion.

We shall denote by s_n the quantities

$$s_n=(\overset{n}{A},A)/(A,A)\ , \tag{1·12}$$

where $\overset{n}{A}=[d^nA(t)/dt^n]_{t=0}$. Although the lowest time derivatives should be written $\overset{1}{A}$, $\overset{2}{A}$, $\cdots$ in this notation, we shall however keep for them the more usual notation $\dot{A}$, $\ddot{A}$, $\cdots$. Now we can develop the relaxation function $\Xi(t)$ into the Taylor series around $t=0$,

$$\Xi(t)=1+\frac{s_1}{1!}t+\frac{s_2}{2!}t^2+\cdots+\frac{s_n}{n!}t^n+\cdots\ . \tag{1·13}$$

Then, ignoring for the time being the question of convergence, we can formally deduce a series expansion for the Laplace transform of $\Xi(t)$, by taking the Laplace transform of each term of the right-hand side of (1·13). We obtain in this way

$$\Xi(z)=\frac{1}{z}+\frac{s_1}{z^2}+\frac{s_2}{z^3}+\cdots+\frac{s_n}{z^{n+1}}+\cdots. \tag{1·14}$$

The series (1·13) and (1·14) are called respectively the moment expansion of the relaxation function and the moment expansion of the Laplace transform of the relaxation function. The reason is that the coefficients s_n are related to the moments μ_n of $\xi(\omega)$, the frequency distribution of $\Xi(t)$, by the equations

$$s_n=\left[\frac{d^n\Xi(t)}{dt^n}\right]_{t=0}=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty}(i\omega)^n\xi(\omega)\,d\omega=i^n\mu_n\,, \tag{1·15}$$

where

$$\xi(\omega)=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty}\Xi(t)e^{-i\omega t}dt\,. \tag{1·16}$$

In connection with the definition of the quantities s_n, we would like to also recall relations existing between the correlation functions of the initial values of the time derivatives of $A(t)$. By simply making a repeated use of the fact that the inner product has the Liouville operator Hermitian and by remembering that $\overset{n}{A}\equiv(iL)^nA$, we obtain for any m and p

$$(\overset{m}{A},\overset{m+2p}{A})=(-1)^p(\overset{m+p}{A},\overset{m+p}{A})=(-1)^m s_{2(m+p)}\,, \tag{1·17}$$

$$\begin{aligned}(\overset{m}{A},\overset{m+2p+1}{A})&=(-1)^p(\overset{m+p}{A},\overset{m+p+1}{A})=(-1)^{p+1}(\overset{m+p+1}{A},\overset{m+p}{A})\\&=(-1)^{m+1}s_{2(m+p)+1}\,.\end{aligned} \tag{1·18}$$

Thus we see that the correlation functions of the form $(\overset{m}{A},\overset{m+2p}{A})$ have real values; on the contrary, correlation functions of the form $(\overset{m}{A},\overset{m+2p+1}{A})$ have pure imaginary values, since

$$(\overset{m+p}{A},\overset{m+p+1}{A})=-(\overset{m+p+1}{A},\overset{m+p}{A})=-(\overset{m+p}{A},\overset{m+p+1}{A})^*\,, \tag{1·19}$$

unless A is an Hermitian variable, in which case they are equal to zero. These last remarks as well as the relations (1·17) and (1·18) will be useful later.

Thus, as we said before, we shall mainly investigate the convergence of the continued fraction (1·11) and the power series (1·14): for this reason, we shall agree throughout this paper, for the sake of brevity, that we are referring to (1·14), when speaking of "the moment expansion". However, the convergence of the moment expansion (1·13) will be also discussed, because beyond the convergence problem in itself, a fundamental problem we wish to approach is to find any general properties of time autocorrelation functions, valid if not for any dynamical variable and any system, at least for large classes of dynamical variables and systems.

Now, for the convergence problem as well as for the general problem of the properties of time autocorrelation functions, as it will be clear later, the sets of quantities s_n on the one hand, and ω_j and Δ_j^2 on the other hand, play symmetric roles, so that it is necessary to know the relation between the two: this will be accomplished in § 2. Whereas the continued fraction expansion was introduced by Mori in the frame of a Brownian motion theory of time relaxation, we shall introduce it from another point of view, which consists in looking for an analytic continuation to the possibly divergent moment expansion. Indeed it is a common practice in Analysis to try to find an analytic continuation to a divergent series by expanding it into a continued fraction, and we shall apply to our physical problem well-known mathematical techniques developed to solve this problem. However, the formalism developed in § 2 will provide us with more than the relation between the two expansions. Actually, as was already pointed out by Mori,[2] the generalized Brownian motion formulation does not depend upon the explicit form of the inner product. Thus we can suspect that we are in presence of a rather general mathematical structure: another purpose of § 2 will be to elucidate this structure. Doing so, we shall find another way of constructing the random forces and identify mathematically some other important physical quantities.

In § 3 we shall discuss the same problem of the relation between the two expansions for the case of Hermitian dynamical variables. The main reason for this special discussion is that it will provide us with formulae important for practical applications.

Then, in § 4, we shall discuss in general the convergence rules of the moment and continued fraction expansions and this general discussion will be illustrated in § 5 by an exactly solvable case, that of an isotopic impurity in a linear chain of coupled harmonic oscillators. Finally, § 6 will be devoted to the question of knowing whether or not the continued fraction expansion might be convergent for any Hermitian dynamical variable and any system, with a discussion of the consequences of such an affirmative answer.*)

§ 2. Relation between the moment expansion and the continued fraction expansion: Case of a general dynamical variable

As we have stated in the previous paragraph, the problem of relating the moment expansion (1·14) to the continued fraction representation (1·11) simply belongs to the general problem of expanding a series into a continued fraction or vice-versa. Such a problem has been treated by mathematicians for a very long time since Frobenius[6],[7] and Stieltjes[8] and it is known at least since Tchebychef[9] that it is related to the problem of constructing a set of polynomials

*) A preliminary report of some of the results presented in this paper was published by the author in Prog. Theor. Phys. **35** (1966), 752.

orthogonal with a given sequence of numbers, namely here the coefficients of the series. In turn, these orthogonal polynomials appear also when trying to establish a correspondence between a certain sequence of real numbers and a certain Jacobi matrix. Thus the mathematical theory has many ramifications and presents much interest by itself. As this formalism may not be so familiar to physicists, we shall first recall some mathematical results, before applying them to the physical problem: in doing so, we shall of course restrict ourselves to the mathematical background indispensable for our purpose; besides, the fact that we start from a sequence of numbers not necessarily real brings some limitation to the possible mathematical developments. With the necessary adaptations, we follow closely the elegant presentation of Akhiezer,[10] to which we refer the reader for proofs and further details.

We then consider an infinite sequence of numbers $s_0=1$, s_1, s_2, $\cdots$, s_n, $\cdots$ which may be complex or real and for which we assume that the Hankel determinants

$$D_n=\begin{vmatrix} s_0 & s_1 & \cdots & s_n \\ s_1 & s_2 & \cdots & s_{n+1} \\ \cdots & \cdots & \cdots & \cdots \\ s_n & s_{n+1} & \cdots & s_{2n} \end{vmatrix} \tag{2·1}$$

are different from zero. To this sequence, we associate a functional $\mathcal{F}$ defined in the space of polynomials by

$$\mathcal{F}\{\alpha_0+\alpha_1\lambda+\alpha_2\lambda^2+\cdots+\alpha_n\lambda^n\}$$
$$=\alpha_0 s_0+\alpha_1 s_1+\alpha_2 s_2+\cdots+\alpha_n s_n\,, \tag{2·2}$$

where α_0, α_1, $\cdots$, α_n are the coefficients of some arbitrary polynomial in λ of degree n. Now, the polynomials $P_n(\lambda)$, where the index n refers to the degree of the polynomial, orthogonal with respect to the sequence s_k, are defined by the condition that

$$\mathcal{F}\{P_n(\lambda)\ P_m(\lambda)\}\quad \begin{cases} =0 & \text{if } m\neq n\,, \\ \neq 0 & \text{if } m=n\,. \end{cases} \tag{2·3}$$

These polynomials can be constructed, the on the one hand, by simply orthogonalizing the sequence of functions $\lambda^k (k=0, 1, 2, \cdots)$ by the usual Schmidt process, taking for scalar product, in view of (2·2),

$$(\lambda^j, \lambda^k)=s_{j+k}\,. \tag{2·4}$$

But, on the other hand, one can also show directly that they are given by the expressions*)

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*) As the definition (2·3) does not specify the normalization of the polynomials $P_n(\lambda)$, we choose the proportionality factor in view of future use. Furthermore, if we agree to put $D_{-1}=1$, the determinantal expression (2·5) remains valid for $n=0$.

$$P_0(\lambda)=1,\ P_n(\lambda)=\frac{1}{D_{n-1}}\begin{vmatrix} s_0 & s_1 & \cdots & s_n \\ s_1 & s_2 & \cdots & s_{n+1} \\ \cdots & \cdots & \cdots & \cdots \\ s_{n-1} & s_n & \cdots & s_{2n-1} \\ 1 & \lambda & \cdots & \lambda^n \end{vmatrix}\quad (n\geq 1). \tag{2·5}$$

Indeed, the polynomials defined by (2·5) are of the form $P_n(\lambda)=\lambda^n+R_{n-1}(\lambda)$, where $R_{n-1}(\lambda)$ is a polynomial of degree $n-1$. Therefore they satisfy (2·3) if $\mathcal{F}\{P_n(\lambda)\lambda^m\}=0$ for $m<n$ and $\mathcal{F}\{P_n(\lambda)\lambda^m\}\neq 0$ for $m=n$. But this is precisely the case for the polynomials (2·5) since

$$\mathcal{F}\{P_n(\lambda)\lambda^m\}=\frac{1}{D_{n-1}}\mathcal{F}\left\{\begin{vmatrix} s_0 & s_1 & \cdots & s_n \\ s_1 & s_2 & \cdots & s_{n+1} \\ \cdots & \cdots & \cdots & \cdots \\ s_{n-1} & s_n & \cdots & s_{2n-1} \\ \lambda^m & \lambda^{m+1} & \cdots & \lambda^{m+n} \end{vmatrix}\right\}=\begin{cases} 0 & \text{if } m<n, \quad (2·6) \\ \dfrac{D_n}{D_{n-1}} & \text{if } m=n. \quad (2·7)\end{cases}$$

A recurrence equation for the polynomials $P_n(\lambda)$ can be easily obtained by expanding $\lambda P_n(\lambda)$ in terms of the polynomials $P_k(\lambda)$ of degree $k\leq n+1$. Making use of (2·3), (2·5), (2·6) and (2·7), we are led to the second order recurrence equation

$$y_{n+1}(\lambda)=(\lambda-a_n)y_n(\lambda)-b_n^2 y_{n-1}(\lambda)\ , \tag{2·8}$$

where

$$a_n=\frac{D_{n-1}}{D_n}\mathcal{F}\{\lambda P_n(\lambda)P_n(\lambda)\}\ , \tag{2·9}$$

$$b_0=0\ ,\qquad b_n^2=\frac{D_n D_{n-2}}{D_{n-1}^2} \tag{2·10}$$

with the initial conditions

$$P_0(\lambda)=1\ ,\qquad P_1(\lambda)=\lambda-a_0\ . \tag{2·11}$$

But another solution of the recurrence equation (2·8) is provided by the polynomials $Q_n(\lambda)$ of degree $n-1$, for which the initial conditions are

$$Q_0(\lambda)=0\ ,\qquad Q_1(\lambda)=1 \tag{2·12}$$

and it can be shown[10),11)] that

$$\frac{Q_n(z)}{P_n(z)}=\frac{s_0}{z}+\frac{s_1}{z^2}+\cdots+\frac{s_{2n-1}}{z^{2n}}+O\left(\frac{1}{z^{2n+1}}\right). \tag{2·13}$$

Let us consider now the infinite continued fraction

$$\cfrac{1}{z-a_0-\cfrac{b_1^2}{z-a_1-\cfrac{b_2^2}{z-a_2-\ddots}}} \tag{2·14}$$

for which the n-th approximant with numerator $N_n(z)$ and denominator $M_n(z)$ is defined by

$$\frac{N_n(z)}{M_n(z)}=\cfrac{1}{z-a_0-\cfrac{b_1^2}{z-a_1-\cfrac{\ddots}{\quad -\cfrac{b_{n-1}^2}{z-a_{n-1}}}}} \quad . \quad (n\geq 1) \tag{2·15}$$

One easily verifies that $M_n(z)$ and $N_n(z)$ obey the recurrence equation (2·8) with initial conditions (2·11) and (2·12) respectively. Therefore we may write

$$\frac{N_n(z)}{M_n(z)}=\frac{Q_n(z)}{P_n(z)} \tag{2·16}$$

and Eq. (2·13) shows that (2·14) is the continued fraction expansion of the formal series associated to the sequence s_k, that is

$$\frac{s_0}{z}+\frac{s_1}{z^2}+\cdots+\frac{s_n}{z^{n+1}}+\cdots \tag{2·17}$$

in this sense that the expansion of the n-th approximant of (2·14) in powers of $1/z$ agrees with (2·17) up to the term s_{2n-1}/z^{2n}.

This establishes the fact that expanding a series into a continued fraction is equivalent to constructing the set of polynomials orthogonal with respect to the coefficients of the series, and (2·9) and (2·10) give us the expressions of the coefficients of the continued fraction in terms of those of the series.

At this point we could already simply use (2·9) and (2·10) to solve the problem of expressing the quantities ω_j and Δ_j^2 in terms of the moments, but the fact that the random forces f_j and the polynomials $P_n(\lambda)$ both form orthogonal sets of vectors in their respective spaces, and the similarity between the expressions $\Delta_j^2=(f_j, f_j)/(f_{j-1}, f_{j-1})$ and $b_j^2=\mathcal{T}\{P_j(\lambda)P_j(\lambda)\}/\mathcal{T}\{P_{j-1}(\lambda)P_{j-1}(\lambda)\}$ let us suspect a close connection between the random forces and the polynomials P_k. In order not only to bring it out, but also, more generally, to bring fully out the connection between the mathematical formalism and Mori's formalism, we shall now consider the previous developments from a completely different point of view.

Let us first introduce the polynomials $R_k(\lambda)$ defined by

$$R_k(\lambda)=\left[(-1)^k\frac{D_{k-1}}{D_k}\right]^{1/2}P_k(\lambda)\ . \tag{2·18}$$

From (2·8), (2·9) and (2·10), we find that they satisfy the recurrence equation

$$-ib_n Y_{n-1}(\lambda) + a_n Y_n(\lambda) + ib_{n+1} Y_{n+1}(\lambda) = \lambda Y_n(\lambda) . \tag{2·19}$$

Now together with the initial condition $(a_0-\lambda)R_0(\lambda)+ib_1R_1(\lambda)=0$, it determines the infinite matrix

$$\begin{pmatrix} a_0 & ib_1 & 0 & 0 & 0\cdots\cdots \\ -ib_1 & a_1 & ib_2 & 0 & 0\cdots\cdots \\ 0 & -ib_2 & a_2 & ib_3 & 0\cdots\cdots \\ 0 & 0 & -ib_3 & a_3 & ib_4\cdots\cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots \end{pmatrix} . \tag{2·20}$$

Following Akhiezer,[10] let us consider this matrix as the representation of a certain linear operator $\mathcal{L}$ in a Hilbert space $\mathcal{H}$. Let us denote by x the vectors of that Hilbert space and by (x_j, x_k) the inner product of x_j by the Hermitian conjugate of x_k.

Taking an orthonormal basis e_k in $\mathcal{H}$, we can first define the operator $\mathcal{L}$ for the unit vectors e_k by the equation

$$\mathcal{L} e_k = -ib_k e_{k-1} + a_k e_k + ib_{k+1} e_{k+1} . \tag{2·21}$$

Since $\mathcal{L}$ is linear, it is also defined by (2·21) for all finite vectors of $\mathcal{H}$. Furthermore, the definition (2·21) determines also every integer non-negative power of the operator $\mathcal{L}$ and therefore every polynomial of the operator $\mathcal{L}$. As the construction of e_k from e_0 requires the same algebraic operations as constructing $R_k(\lambda)$ from $R_0(\lambda)=1$, we may write

$$e_k = [R_k(\mathcal{L})]e_0 \tag{2·22}$$

and to the expansion

$$\lambda^m = \sum_{j=0}^{m} \alpha_j^{(m)} R_j(\lambda) \tag{2·23}$$

does correspond the expansion

$$\mathcal{L}^m e_0 = \sum_{j=0}^{m} \alpha_j^{(m)} e_j = \sum_{j=0}^{m} \alpha_j^{(m)} [R_j(\mathcal{L})]e_0 . \tag{2·24}$$

By applying the functional $\mathcal{F}$ on (2·23), we then obtain

$$\mathcal{F}(\lambda^{m+n}) = s_{m+n} = \sum_{j=0}^{m} \sum_{k=0}^{n} \alpha_j^{(m)} \alpha_k^{(n)} \delta_{jk} , \tag{2·25}$$

whereas from (2·24), we deduce

$$(\mathcal{L}^m e_0, \mathcal{L}^n e_0) = \sum_{j=0}^{m} \sum_{k=0}^{n} \alpha_j^{(m)} \alpha_k^{(n)*} \delta_{jk} . \tag{2·26}$$

We now are in the position to apply the previous mathematical formalism to our physical problem.

We wish to relate (1·11) to (1·14) in the same way as we have related (2·14) to (2·17). Now, in (1·11) the coefficients $a_n = i\omega_n$ are pure imaginary quantities: indeed, using (1·7), one has

$$\frac{(\dot{f}_j, f_j)}{(f_j, f_j)} = \frac{(iLf_j, f_j)}{(f_j, f_j)} = -\frac{(f_j, iLf_j)}{(f_j, f_j)} = -\frac{(iLf_j, f_j)^*}{(f_j, f_j)} = -\frac{(\dot{f}_j, f_j)^*}{(f_j, f_j)} . \tag{2·27}$$

Thus it follows from (2·21) that

$$(\mathcal{L}e_k, e_k) = a_k = -a_k^* = -(e_k, \mathcal{L}e_k) . \tag{2·28}$$

On the other hand, the coefficients $\Delta_n^2 = -b_n^2$ are real and positive numbers. Therefore one has

$$\left.\begin{aligned} (\mathcal{L}e_k, e_{k-1}) &= ib_k = -(e_k, \mathcal{L}e_{k-1}) , \\ (\mathcal{L}e_k, e_{k+1}) &= -ib_k = -(e_k, \mathcal{L}e_{k+1}) . \end{aligned}\right\} \tag{2·29}$$

From (2·28), (2·29) and (2·21) we can conclude, for any vectors e_j and e_k and, more generally since $\mathcal{L}$ is linear, for any finite vectors x_j and x_k, that we have the equalities

$$\left.\begin{aligned} (\mathcal{L}e_j, e_k) &= -(e_j, \mathcal{L}e_k) , \\ (\mathcal{L}x_j, x_k) &= -(x_j, \mathcal{L}x_k) . \end{aligned}\right\} \tag{2·30}$$

These equalities lead to

$$\mathcal{L} = -\mathcal{L}^{\dagger} \tag{2·31}$$

and Eq. (2·26) yields then

$$(\mathcal{L}^m e_0, \mathcal{L}^n e_0) = \sum_{j=0}^{m} \sum_{k=0}^{n} (-1)^n \alpha_j^{(m)} \alpha_k^{(n)} \delta_{jk} = (-1)^n s_{m+n} . \tag{2·32}$$

Now let us compare this last equation with (1·17) and (1·18). Remembering that $\overset{n}{A} \equiv (iL)^n A$, we immediately recognize that in the relaxation function expansion problem, we have

$$\mathcal{L} \equiv iL , \qquad e_0 \equiv A \tag{2·33}$$

and that generally, *we can deduce the quantities appearing in Mori's formalism from those appearing in the orthogonal polynomials formalism just by replacing* λ *by* iL *and then by operating on* A *with the resulting function of the operator* iL.

If we perform this operation on $P_k(\lambda)$, we obtain the random force f_k:

$$f_k \equiv [P_k(iL)] A . \tag{2·34}$$

Although this relation is fairly obvious from the properties of the two sets of quantities, one can reason in the following way. On the one hand, the polynomials $P_k(\lambda)$ can be constructed by a Schmidt orthogonalization of the sequence of functions λ^k, $k=0, 1, 2\cdots$. On the other hand, one can see from Mori's definition of the random forces, that *the random forces can be obtained by orthogonalizing the sequence of time derivatives* $\overset{k}{A}$, $k=0, 1, 2, \cdots$ *according to*

$$f_n = \overset{n}{A} - \sum_{l=0}^{n-1} \frac{(\overset{n}{A}, f_l)}{(f_l, f_l)} f_l . \tag{2·35}$$

One has just to notice that the recurrence equation leads to an expression of f_n linear with respect to the $\overset{k}{A}(k\leq n)$, where the coefficient of the highest order time derivative is unity. The resulting linear system can be solved to give an expression of $\overset{n}{A}$ linear with respect to the vectors $f_k(k\leq n)$, leading to (2·35) after the orthogonality of the vectors f_k has been taken into account. Thus, since one goes from the sequence λ^k to the sequence $\overset{k}{A}$ by the above mentioned correspondence rule, the identity (2·34) is proved. By using (2·5), one can then write for f_n the expression

$$f_n = \frac{1}{D_{n-1}} \begin{vmatrix} s_0 & s_1 & \cdots & s_n \\ s_1 & s_2 & \cdots & s_{n+1} \\ \cdots & \cdots & \cdots & \cdots \\ s_{n-1} & s_n & \cdots & s_{2n-1} \\ A & \dot{A} & \cdots & \overset{n}{A} \end{vmatrix} \tag{2·36}$$

and from (2·3) and (2·6) the static correlation functions of f_n are seen to be given by

$$(f_n, f_m) = (f_n, \overset{m}{A}) = \begin{cases} 0 & \text{if } m<n , \\ (-1)^n \dfrac{D_n}{D_{n-1}} (A, A) & \text{if } m=n . \end{cases} \tag{2·37}$$

From (2·9) and (2·10) follow the expressions of the coefficients $i\omega_n$ and Δ_n^2 in terms of the moments:

$$i\omega_n = \frac{1}{D_n D_{n-1}} \left(\begin{vmatrix} s_0 & s_1 & \cdots & s_n \\ s_1 & s_2 & \cdots & s_{n+1} \\ \cdots & \cdots & \cdots & \cdots \\ s_{n-1} & s_n & \cdots & s_{2n-1} \\ \dot{A} & \ddot{A} & \cdots & \overset{n+1}{A} \end{vmatrix} , \begin{vmatrix} s_0 & s_1 & \cdots & s_n \\ s_1 & s_2 & \cdots & s_{n+1} \\ \cdots & \cdots & \cdots & \cdots \\ s_{n-1} & s_n & \cdots & s_{2n-1} \\ A & \dot{A} & \cdots & \overset{n}{A} \end{vmatrix} \right) , \tag{2·38}$$

$$\Delta_n{}^2 = -\frac{D_{n-2}D_n}{D_{n-1}^2}. \tag{2·39}$$

As for the recurrence equation which is satisfied by the random forces, it is readily deduced from (2·8),

$$f_n = \left(iL - \frac{(iLf_{n-1}, f_{n-1})}{(f_{n-1}, f_{n-1})}\right) f_{n-1} + \Delta^2{}_{n-1} f_{n-2} \tag{2·40}$$

and can also be written

$$f_n = \left(iL - \frac{(\dot{f}_{n-1}, f_{n-1})}{(f_{n-1}, f_{n-1})}\right) f_{n-1} + \Delta^2{}_{n-1} f_{n-2} \tag{2·41}$$

as it is seen from (1·7). Equation (2·40) is easily seen to coincide with Mori's definition (1·3), once the orthogonality of the random forces has been taken into account. Indeed, (2·40) yields

$$(iLf_{n-1}, f_l) = 0 \qquad \text{if} \quad l \leq n-3, \tag{2·42}$$

$$(iLf_{n-1}, f_{n-2}) = -\Delta_{n-1}^2 (f_{n-2}, f_{n-2}) = -(f_{n-1}, f_{n-1}), \tag{2·43}$$

which shows that Mori's recurrence equation reduces to (2·40).

Finally let us mention that the representation of the operator iL in the base provided by the normalized random forces $f_k/(f_k, f_k)^{1/2}$ is the matrix

$$\begin{pmatrix} i\omega_0 & \Delta_1 & 0 & 0 & 0 & \cdots \\ -\Delta_1 & i\omega_1 & \Delta_2 & 0 & 0 & \cdots \\ 0 & -\Delta_2 & i\omega_2 & \Delta_3 & 0 & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{pmatrix} \tag{2·44}$$

as can be easily verified by computing the matrix elements with help of Eqs. (2·40) and (2·41).

As we have stated in the introduction, the fact that Mori's formalism did not depend upon the explicit form of the inner product, let us suspect that we were in presence of a fairly general structure: this structure is now elucidated. The most interesting result is perhaps represented by the identity (2·34) or, equivalently, by the fact that the random forces may be constructed by a Schmidt orthogonalization of the set of time derivatives $\overset{k}{A}$. Furthermore, from a more practical point of view, we have obtained expressions for the coefficients of the continued fraction expansion in terms of the moments. The inverse problem, that is to express the coefficients of the series in terms of those of the continued fraction, was solved a long time ago by Stieltjes,[8),12)] using a completely different method. As the resulting expressions are far from being as simple as (2·38) and (2·39), and as we shall not use them later, we shall not reproduce them here. But we shall see in the next section that in the case of an Hermitian variable, a new direct method produces again simple relations.

§ 3. Relation between the moment expansion and the continued fraction expansion: case of an Hermitian variable

In this section we shall treat the same problem as in § 2 for the case of Hermitian dynamical variables: by an Hermitian variable, we mean a real phase function in the classical case or an Hermitian operator in the quantum case.

The general formalism developed in the previous section could of course be applied to such variables, and for this reason we shall be somewhat briefer, since some of the results relative to the Hermitian case are very close or even similar to the general ones. However, it is instructive to consider separately the case of Hermitian variables at least for two reasons. First, using a new method, we shall adopt a more physical attitude and take up the problem as it presents itself in the development of the physical theory: namely, we shall start from Mori's definition of the vectors f_j. Secondly, this direct method has the advantage to lead naturally to expressions valid to general order, and which may be useful in practical calculations (Eqs. (3·8)-(3·11)), as it will be demonstrated in § 5. Let us add, from the same practical point of view, that the class of Hermitian variables is very large and contains, in particular, all fluxes associated with transport processes due to thermal, momentum or concentration gradients; however, notable exceptions exist, such as, for example, the normal coordinates of sound waves.[1]

If the dynamical variable $A(t)$ is supposed to be Hermitian, then it follows from (1·19) that every s_j with odd index vanishes,

$$s_{2n+1}=0, \qquad n=0,\ 1,\ 2,\ \cdots, \tag{3·1}$$

whereas s_j with even index, according to (1·17) take the form

$$s_{2n}=(-1)^n(\overset{n}{A},\ \overset{n}{A})/(A,\ A), \qquad n=0,\ 1,\ 2,\ \cdots. \tag{3·2}$$

It is therefore convenient to introduce the positive quantities c_n equal to the moments μ_{2n}, defined by

$$c_0=1, \qquad c_n=(\overset{n}{A},\ \overset{n}{A})/(A,\ A)=(-1)^n s_{2n}=\mu_{2n}, \qquad n\geq 1, \tag{3·3}$$

so that the moment expansion (1·14) reduces to

$$\frac{1}{z}-\frac{c_1}{z^3}+\frac{c_2}{z^5}-\cdots+(-1)^n\frac{c_n}{z^{2n+1}}+\cdots. \tag{3·4}$$

On the other hand, if A is Hermitian, then all the f_j are also Hermitian and (2·27) yields

$$\omega_j=0, \qquad j=0,\ 1,\ 2,\ \cdots. \tag{3·5}$$

Correspondingly, the infinite continued fraction (1·11) reduces to

$$\cfrac{1}{z+\cfrac{\Delta_1^2}{z+\cfrac{\Delta_2^2}{z+\ddots}}}\,. \tag{3·6}$$

Let us now turn to the problem of expressing the coefficients Δ_j^2 in terms of the moments c_k and vice-versa.

We start by applying Mori's recurrence formula (1·3) to the construction of the vectors f_j with lowest indices. Taking into account (3·1) and (3·2) and making appropriate rearrangements, one can bring the expressions of the random forces and their static correlation functions into the following form:

$$\begin{aligned}
&f_1=\dot{A}\,, && (f_1,f_1)/(A,A)=c_1\,,\\
&f_2=\ddot{A}+\Delta_1^2 A\,, && (f_2,f_2)/(A,A)=c_2-\Delta_1^2 c_1\,,\\
&f_3=\overset{3}{A}+(\Delta_1^2+\Delta_2^2)\dot{A}\,, && (f_3,f_3)/(A,A)=c_3-(\Delta_1^2+\Delta_2^2)c_2\,,\\
&f_4=\overset{4}{A}+(\Delta_1^2+\Delta_2^2+\Delta_3^2)\ddot{A}+\Delta_1^2\Delta_3^2 A\,, &&\\
&(f_4,f_4)/(A,A)=c_4-(\Delta_1^2+\Delta_2^2+\Delta_3^2)c_3+\Delta_1^2\Delta_3^2 c_2\,, &&\\
&f_5=\overset{5}{A}+(\Delta_1^2+\Delta_2^2+\Delta_3^2+\Delta_4^2)\overset{3}{A}+(\Delta_1^2\Delta_3^2+\Delta_1^2\Delta_4^2+\Delta_2^2\Delta_4^2)\dot{A}\,, &&\\
&(f_5,f_5)/(A,A)=c_5-(\Delta_1^2+\Delta_2^2+\Delta_3^2+\Delta_4^2)c_4+(\Delta_1^2\Delta_3^2+\Delta_1^2\Delta_4^2+\Delta_2^2\Delta_4^2)c_3\,, &&
\end{aligned} \tag{3·7}$$

where the quantities Δ_j^2 are defined by (1·10).*) We have given the expressions (3·7) because they may be useful for practical calculations and illustrate the general expressions which follow. Indeed a careful examination of (3·7) suggests that these general expressions are

$$\begin{aligned}
f_{2n}=\overset{2n}{A}+S_1^{(2n-1)}\overset{2n-2}{A}+S_2^{(2n-1)}\overset{2n-4}{A}+\cdots+S_p^{(2n-1)}\overset{2n-2p}{A}+\cdots&\\
+S_n^{(2n-1)}A\,,&
\end{aligned} \tag{3·8}$$

$$\begin{aligned}
f_{2n-1}=\overset{2n-1}{A}+S_1^{(2n-2)}\overset{2n-3}{A}+S_2^{(2n-2)}\overset{2n-5}{A}+\cdots+S_p^{(2n-2)}\overset{2n-2p-1}{A}+\cdots&\\
+S_{n-1}^{(2n-2)}\dot{A}\,,&
\end{aligned} \tag{3·9}$$

$$\begin{aligned}
(f_{2n},f_{2n})/(A,A)=c_{2n}-S_1^{(2n-1)}c_{2n-1}+S_2^{(2n-1)}c_{2n-2}-\cdots&\\
+(-1)^p S_p^{(2n-1)}c_{2n-p}+\cdots+(-1)^n S_n^{(2n-1)}c_n\,,&
\end{aligned} \tag{3·10}$$

---

*) In order to obtain these expressions, the following two relations have been useful:

$$(f_2,f_2)/(A,A)=c_2-c_1^2;\quad (f_3,f_3)/(A,A)=c_3-\frac{c_2^2}{c_1}\,.$$

$$(f_{2n-1}, f_{2n-1})/(A, A) = c_{2n-1} - S_1^{(2n-2)} c_{2n-2} + S_2^{(2n-2)} c_{2n-3} - \cdots$$
$$+ (-1)^p S_p^{(2n-2)} c_{2n-p-1} + \cdots + (-1)^{n-1} S_{n-1}^{(2n-2)} c_n . \qquad (3\cdot 11)$$

In these expressions $S_1^{(n)} = \sum_{i=1}^{n} \Delta_i^2$ and $S_p^{(n)}$ denotes the sum of all possible products of p different factors Δ_k^2 chosen in the set $\Delta_1^2, \Delta_2^2, \cdots, \Delta_n^2$ in such a way that all the indices k differ from each other by more than 1. For example, the last two expressions (3·7) show that $S_2^{(4)} = \Delta_1^2\Delta_3^2 + \Delta_1^2\Delta_4^2 + \Delta_2^2\Delta_4^2$. Referring to the definition of Δ_j^2, we see that these products are such that there is no cancellation between autocorrelation functions appearing in the numerator and the denominator.

In order to prove the validity of Eqs. (3·8) and (3·9), we simply have to show that they satisfy the recurrence relations (1·3), or equivalently (2·40). If one notes that the sums $S_p^{(n)}$ obey the recurrence relations

$$S_p^{(n)} = S_p^{(n-1)} + S_{p-1}^{(n-2)} \Delta_n^2 , \qquad (3\cdot 12)$$

then one easily verifies that the expressions (3·8) and (3·9) satisfy the recurrence equation

$$f_n = iLf_{n-1} + \Delta_{n-1}^2 f_{n-2} . \qquad (3\cdot 13)$$

But, if one makes use of (3·5), this recurrence equation is seen to be identical to (2·40). Therefore the expressions (3·8) and (3·9) are established.

The same expressions provide us with a set of equations linear with respect to the vectors $\overset{n}{A}$ and can be solved to yield the vectors $\overset{n}{A}$ in terms of the random forces f_n. The resulting expressions for even order time derivatives are the determinants

$$\overset{2n}{A} = \begin{vmatrix} f_{2n} & S_1^{(2n-1)} & S_2^{(2n-1)} & \cdots & S_n^{(2n-1)} \\ f_{2n-2} & 1 & S_1^{(2n-3)} & \cdots & S_{n-1}^{(2n-3)} \\ f_{2n-4} & 0 & 1 & \cdots & S_{n-2}^{(2n-5)} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ f_0 & 0 & 0 & \cdots & 1 \end{vmatrix} \qquad (3\cdot 14)$$

obtained by considering the $n+1$ equations corresponding to even f_j, $j \leq 2n$. The expressions for time derivatives of odd order are very similar and need not be exhibited. From the fact that the coefficient of f_n is unity and from the orthogonality of the vectors f_n, it follows that

$$(f_n, f_n) = (f_n, \overset{n}{A}) \qquad (3\cdot 15)$$

which proves (3·10) and (3·11), if one takes into account (1·17). Furthermore, we can write the expansion of (3·14) as

$$\overset{n}{A}=f_n+\sum_{l=0}^{n-1}\frac{(\overset{n}{A},f_l)}{(f_l,f_l)}f_l\,, \tag{3·16}$$

whence (2·35) is again deduced, showing here with full detail that the random forces can be constructed by orthogonalizing the sequence of vectors $\overset{n}{A}(n=0,1,2,\cdots)$ by the usual Schmidt process.

We can say that the expressions (3·10) and (3·11) are mixed in the sense that they give the static autocorrelation functions of the random forces in terms of both the moments c_k and the quantities Δ_j^2. Therefore we must go one step further in order to obtain the expressions of the c_k in terms of the Δ_j^2 only and vice-versa.

In order to obtain the expressions of the c_k in terms of the Δ_j^2, we simply have to notice that $(f_n,f_n)/(A,A)=\Delta_n^2\Delta_{n-1}^2\cdots\Delta_1^2$. Thus, the expressions (3·10) and (3·11) considered as equations linear with respect to the moments can be solved immediately to yield

$$c_{2n}=\begin{vmatrix} \prod_{i=1}^{2n}\Delta_i^2 & -S_1^{(2n-1)} & S_2^{(2n-1)}\cdots(-1)^nS_n^{(2n-1)} & 0 & \cdots 0 & 0 \\ \prod_{i=1}^{2n-1}\Delta_i^2 & 1 & -S_1^{(2n-2)}\cdots(-1)^{n-1}S_{n-1}^{(2n-2)} & 0 & \cdots 0 & 0 \\ \prod_{i=1}^{2n-2}\Delta_i^2 & 0 & 1\quad\cdots(-1)^{n-2}S_{n-2}^{(2n-3)} & (-1)^{n-1}S_{n-1}^{(2n-3)}\cdots & 0 & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ \Delta_1^2\Delta_2^2 & 0 & 0\quad\cdots\quad 0 & 0 & \cdots 1 & -\Delta_1^2 \\ \Delta_1^2 & 0 & 0\quad\cdots\quad 0 & 0 & \cdots 0 & 1 \end{vmatrix} \tag{3·17}$$

and

$$c_{2n-1}=\begin{vmatrix} \prod_{i=1}^{2n-1}\Delta_i^2 & -S_1^{(2n-2)} & S_2^{(2n-2)}\cdots(-1)^{n-1}S_{n-1}^{(2n-2)} & 0 & \cdots 0 & 0 \\ \prod_{i=1}^{2n-2}\Delta_i^2 & 1 & -S_1^{(2n-3)}\cdots(-1)^{n-2}S_{n-2}^{(2n-3)} & (-1)^{n-1}S_{n-1}^{(2n-3)}\cdots & 0 & 0 \\ \prod_{i=1}^{2n-3}\Delta_i^2 & 0 & 1\quad\cdots(-1)^{n-3}S_{n-3}^{(2n-4)} & (-1)^{n-2}S_{n-2}^{(2n-4)}\cdots & 0 & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ \Delta_1^2\Delta_2^2 & 0 & 0\quad\cdots\quad 0 & 0 & \cdots 1 & -\Delta_1^2 \\ \Delta_1^2 & 0 & 0\quad\cdots\quad 0 & 0 & \cdots 0 & 1 \end{vmatrix}. \tag{3·18}$$

Reciprocally, in order to obtain the expressions of the coefficients Δ_j^2 in terms of the c_k, we first note that in view of Eq. (3·16), one has

$$(f_n,\overset{k}{A})=0 \qquad \text{if} \quad k<n\,. \tag{3·19}$$

Then let us combine the n equations (3·19) corresponding to $k=0, 2, 4, \cdots, 2n-2$ with Eq. (3·10). We have a system of $n+1$ equations linear with respect to the n variables $S_1^{(2n-1)}, S_2^{(2n-1)}, \cdots, S_n^{(2n-1)}$. If we introduce the Hankel determinants B_n defined by

$$B_0=B_1=1, \qquad B_n=\begin{vmatrix} 1 & c_1 & c_2 & \cdots & c_{n-1} \\ c_1 & c_2 & c_3 & \cdots & c_n \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ c_{n-1} & c_n & c_{n+1} & \cdots & c_{2n-2} \end{vmatrix} \quad (n>1), \qquad (3\cdot 20)$$

the condition of compatibility yields

$$(f_{2n}, f_{2n})/(A, A)=B_{n+1}/B_n\,. \qquad (3\cdot 21)$$

In the same way, let us combine the $n-1$ equations (3·19) corresponding to $k=1, 3, \cdots, 2n-3$ with Eq. (3·11). Introducing the other Hankel determinant C_n defined by

$$C_0=1, \qquad C_n=\begin{vmatrix} c_1 & c_2 & c_3 & \cdots & c_n \\ c_2 & c_3 & c_4 & \cdots & c_{n+1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ c_n & c_{n+1} & c_{n+2} & \cdots & c_{2n-1} \end{vmatrix} \quad (n\geq 1), \qquad (3\cdot 22)$$

we obtain through the compatibility condition

$$(f_{2n-1}, f_{2n-1})/(A, A)=C_n/C_{n-1}\,. \qquad (3\cdot 23)$$

The combination of (3·21) and (3·23) finally leads to the relations which we were seeking:

$$\Delta_{2n}^2=\frac{B_{n+1}}{B_n}\frac{C_{n-1}}{C_n}, \qquad \Delta_{2n-1}^2=\frac{B_{n-1}}{B_n}\frac{C_n}{C_{n-1}}. \qquad (3\cdot 24)$$

These relations could of course have been deduced from (2·39) by putting equal to zero all the s_j with odd index and then making suitable interchanges of rows and columns.

We then have completely solved the problem of relating the moment expansion to the continued fraction expansion. The method developed for the Hermitian case may appear simple compared to the mathematical machinery used in § 2, and one may wonder if this direct method could not be extended to the general case: the reason is that in the general case, the non-validity of (3·1) and (3·2) complicates the calculations and precludes the existence of expressions as simple as (3·8)-(3·11), the obtention of which is the key of the method.

We shall close this section by mentioning some interesting inequalities arising from (3·24). Indeed the expressions (3·24) show that all the determinants B_n are of the same sign and that all the determinants C_n are also of the same sign. Since $B_0 = C_0 = 1$, all these determinants must be positive. But it is well known[13] that in this case, all Hankel determinants of the form

$$\begin{vmatrix} c_p & c_{p+1} & \cdots & c_{p+q} \\ c_{p+1} & c_{p+2} & \cdots & c_{p+q+1} \\ \cdots & \cdots & \cdots & \cdots \\ c_{p+q} & c_{p+q+1} & \cdots & c_{p+2q} \end{vmatrix}, \tag{3·25}$$

where p and q are arbitrary, are positive. In particular

$$\begin{vmatrix} c_n & c_{n+1} \\ c_{n+1} & c_{n+2} \end{vmatrix} > 0 \tag{3·26}$$

or

$$\frac{c_{n+2}}{c_{n+1}} > \frac{c_{n+1}}{c_n}, \tag{3·27}$$

that is, the ratio c_{n+1}/c_n always increases with n. This remark will be important in the next section. It should be added, however, that we can also derive the inequality (3·27) directly by applying the Schwarz inequality to the inner product $(\overset{n+2}{A}, \overset{n}{A})$:

$$(\overset{n+2}{A}, \overset{n}{A})^2 = (\overset{n+1}{A}, \overset{n+1}{A})^2 \leq (\overset{n+2}{A}, \overset{n+2}{A})(\overset{n}{A}, \overset{n}{A}). \tag{3·28}$$

But the remark concerning the sign of the determinants B_n and C_n will be useful in the next section.

§ 4. Convergence criteria of the moment and continued fraction expansions

In this section we shall confine ourselves to Hermitian dynamical variables. The reason for this is that in this case, the infinite continued fraction (1·11) takes the form

$$J(z) = \cfrac{1}{z + \cfrac{\Delta_1^2}{z + \cfrac{\Delta_2^2}{z + \ddots}}} = \cfrac{z}{z^2 + \cfrac{\Delta_1^2}{1 + \cfrac{\Delta_2^2}{z^2 + \ddots}}} \tag{4·1}$$

and thus $z^{-1}J(z)$ is a Stieltjes-type continued fraction with respect to the variable $z^2 = u$; now the analytic properties of Stieltjes continued fractions are much simpler to study than that of general continued fractions with complex coefficients.

In view of (4·1) it will be convenient to introduce the infinite continued fraction

$$J(u)\equiv z^{-1}J(z)=\cfrac{1}{u+\cfrac{\varDelta_1^2}{1+\cfrac{\varDelta_2^2}{u+\ddots}}}\tag{4·2}$$

In the same way, to the moment expansion (1·14), which we denote by $S(z)$, corresponds the series expansion

$$S(u)\equiv z^{-1}S(z)=\frac{1}{u}-\frac{c_1}{u^2}+\frac{c_2}{u^3}-\cdots+(-1)^n\frac{c_n}{u^{n+1}}+\cdots.\tag{4·3}$$

We have introduced the symbols J and S, to insist on the fact that both expansions are *representations* of $\Xi(z)$ in the complex plane, with restricted domains of validity: as we pointed out earlier, *the continued fraction may possibly play the role of an analytic continuation for the moment expansion.* As a matter of fact, we note that $J(z)$ and $S(z)$ have the symmetry $J(z)=-J(-z)$, $S(z)=-S(-z)$, whereas $\Xi(z)\neq-\Xi(-z)$. The reason is easy to understand both mathematically and physically. Let us first take the physical point of view and choose for inner product Kubo's canonical product

$$(A,B)=\frac{1}{\beta}\int_0^\beta\langle e^{\lambda H}Ae^{-\lambda H}B^\dagger\rangle d\lambda\,,\tag{4·4}$$

where $\beta=(kT)^{-1}$, T being the absolute temperature and k Boltzmann's constant, and the brackets denote the canonical average. We have, in the quantum case,

$$\begin{aligned}(A(z),A)&=\int_0^\infty dt\exp(-zt)\sum_l\sum_{l'}\exp(i(E_l-E_{l'})t/\hbar)A_{ll'}A^\dagger_{l'l}\exp(-\beta E_l)\\&=\sum_l\sum_{l'}\frac{1}{z-i(E_l-E_{l'})/\hbar}A_{ll'}A^\dagger_{l'l}\exp(-\beta E_l)\,,\end{aligned}\tag{4·5}$$

where E_l is an eigenvalue of the Hamiltonian and $A_{ll'}$ a matrix element of A in the Hamiltonian base. Thus $\Xi(z)$ has poles located on the imaginary axis, and when the system becomes infinitely large, these poles will form a singular line limited by branch points located either at finite distance or at infinity.

On the other hand, from the mathematical point of view, it is well known that all the poles of the approximants of the Stieltjes continued fraction $J(u)$ are located on the negative part of the real axis of the u-plane. To these poles correspond singularities on the imaginary axis of the z-plane. Therefore, all criteria ensuring convergence of $J(u)$ out of the negative part of the real axis, will ensure that $J(z)$ represents $\Xi(z)$ only for z such that $\mathrm{Re}\, z>0$, and in order to continue into the half-plane $\mathrm{Re}\, z<0$ the definition of the function to which $J(z)$ converges, one will have to introduce a Riemann surface and find the physically meaningful Riemann sheet.*)

Keeping these remarks in mind, let us turn to the convergence criteria of $J(u)$ and $S(u)$. As we have emphasized it in § 2, the sets of quantities c_k on the one hand and Δ_j^2 on the other hand, play equivalent roles, so that it is necessary to have criteria expressed in terms of either set for both expansions. Indeed, there may be physical situations in which the moments have simple expressions, whereas the quantities Δ_j^2 are of a very complicated form or even are impossible to obtain to general order. Actually, in view of the results obtained in § 3, it is that situation which one would normally expect. Once $A(t)$ is given, the moments are, in principle, directly accessible by repeated time derivation and scalar product formation, whereas the evaluation of the quantities Δ_j^2 requires more calculations. However, the inverse situation, where the Δ_j^2 have simple expressions, whereas the moments are given by complicated formulae, may also occur, as will be shown in § 5.

We first consider $S(u)$ and $S(z)$. We have seen at the end of § 4 that the ratio c_{n+1}/c_n increases with the index n. Therefore two cases may happen:

a) The ratio c_{n+1}/c_n has an upper limit l:

$$\lim_{n\to\infty} c_{n+1}/c_n = l\,. \tag{4·6}$$

Thus $S(u)$ is convergent for $|u|>l$, divergent for $|u|\leq l$ and $S(z)$ represents $\Xi(z)$ for $|z|>\sqrt{l}$, with $\mathrm{Re}\, z>0$.

b) The ratio c_{n+1}/c_n increases without limit: thus $S(u)$ and $S(z)$ are always divergent, except at infinity. They are asymptotic series.

If we now wish to have criteria in terms of the coefficients Δ_j^2, we can use a result due to Stieltjes.[14] This result states that case a happens if the quantities Δ_n^2 have an upper limit:

$$\lim_{n\to\infty} \Delta_n^2 = \Delta^2 \tag{4·7}$$

and that in such a case, one has the inequality

$$l < 4\Delta^2\,. \tag{4·8}$$

On the contrary, if the quantities Δ_n^2 have no upper limit, then case b happens.

Furthermore, analogous results hold for the largest root of the polynomial $P_n(z)$, which we shall denote by z_n. In case a, one has $|z_n|<\sqrt{l}$ and in case b, z_n goes to infinity with n.

*) The author is indebted to Professor H. Mori for this last remark and for pointing out to him Eq. (4·5).

Next, let us consider $J(u)$ and $J(z)$. We introduce the numbers k_n defined by the recurrence law

$$k_1=1\,, \qquad k_n k_{n+1}=\frac{1}{\Delta_n^2} \tag{4·9}$$

leading to

$$k_{2p}=\frac{\Delta_2^2}{\Delta_1^2}\frac{\Delta_4^2}{\Delta_3^2}\cdots\frac{\Delta_{2p-2}^2}{\Delta_{2p-3}^2}\frac{1}{\Delta_{2p-1}^2}\,;\quad k_{2p+1}=\frac{\Delta_1^2}{\Delta_2^2}\frac{\Delta_3^2}{\Delta_4^2}\cdots\frac{\Delta_{2p-1}^2}{\Delta_{2p}^2}\,,\quad (p\geq 1), \tag{4·10}$$

so that the infinite continued fraction $J(z)$ takes the form

$$J(z)=\cfrac{1}{k_1 z+\cfrac{1}{k_2 z+\cfrac{1}{k_3 z+\ddots}}}\,. \tag{4·11}$$

Then we can apply a theorem due to Stieltjes,[14] which says that if the series $\sum_p k_p$ is divergent, $J(u)$ is uniformly convergent over every finite closed domain of the complex u-plane, the distance of which from the negative half of the real axis is positive; furthermore, the value of $J(u)$ is an holomorphic function of u for all u not on the negative part of the real axis. It follows then that under the same condition, and for every z such that $\mathrm{Re}\, z>0$, $J(z)$ represents the relaxation function $\Xi(z)$, holomorphic in that domain.

If we wish, on the other hand to have a criterion in terms of the moments c_k, we may use a theorem derived by Carleman in the theory of quasi-analytic functions.[15] This theorem states that if the determinants B_n and C_n are positive, and if the series $\sum_n c_n^{-1/2n}$ is divergent, then the Stieltjes moment problem is determinate. But it is well known that the Stieltjes moment problem is determinate if and only if the positive term series $\sum_p k_p$ is divergent. Therefore we may conclude that if the series $\sum_n c_n^{-1/2n}$ is divergent, then $J(u)$ converges to an holomorphic function of u for all u not on the negative part of the real axis; the same conclusion as above follows for $J(z)$. We should like to remark at this point that although the moment problem has necessarily a solution in the time autocorrelation function case—indeed the coefficients c_k are not any numbers, but moments, and it was shown at the end of § 3 that the determinants B_n and C_n are positive as required—nothing proves a priori that this solution is unique.

It should be kept in mind that the two criteria given are of course not unique for deciding about the convergence of $J(u)$ or $J(z)$, but they have seemed to us most appropriate for our purpose. Before discussing their implications, we shall first show on an example which can be exactly solved, how the general mathematical theory of the last three sections can be practically applied.

§ 5. An example: the momentum autocorrelation function of an isotopic impurity in a linear chain of coupled harmonic oscillators

As an illustration of the previous mathematical developments, we shall study the momentum autocorrelation function of an isotopic impurity in a linear chain of coupled harmonic oscillators.

We consider a linear chain of atoms of mass M oscillating harmonically around sites labelled $-N$, $-(N-1)$, $\cdots$, -2 -1, 1, 2, $\cdots$, $N-1$, N, with an isotopic atom of mass $M'=M(1+Q)$ located at the center site 0. The atoms are coupled by springs of force constant K and the atoms at both ends of the chain are connected by springs to fixed walls.

Such a model has been studied extensively by many authors from various points of view, either with respect to irreversibility by Hemmer,[16] Rubin,[17] Takeno and Hori,[18] Turner,[19] or with respect to the theory of Brownian motion by Ullersma.[20]

If u_j is the displacement of the j-th atom, it is convenient to introduce the coordinates $x_j'=\sqrt{M}\,u_j(j\neq 0)$, $x'=\sqrt{M'}\,u_0$ and to define the momenta by $p_j'=\sqrt{M}\,\dot{u}_j(j\neq 0)$, $p'=\sqrt{M'}\,\dot{u}_0$ so that $p_j'=\dot{x}_j'$, $p_0'=\dot{x}'$. Then, if one introduces the normal coordinates

$$\left.\begin{aligned}
&x_s=\sum_{j=-1}^{-N}x_j'\sqrt{\frac{2}{N+1}}\sin\frac{\pi j}{N+1}s\,, && -N\leq s\leq -1\\
&x_s=\sum_{j=1}^{N}x_j'\sqrt{\frac{2}{N+1}}\sin\frac{\pi j}{N+1}s\,, && 1\leq s\leq N\\
&x=x'\,,\\
&p_s=\dot{x}_s\,, \qquad p=\dot{x}\,,
\end{aligned}\right\}\tag{5·1}$$

the Hamiltonian takes the form

$$H=\frac{p^2}{2}+\frac{\omega_0{}^2x^2}{2}+\sum_s\left(\frac{p_s{}^2}{2}+\frac{\omega_s{}^2x_s{}^2}{2}\right)+\omega_0\sum_s A_sx_sx\,,\tag{5·2}$$

where

$$\left.\begin{aligned}
&\omega_0{}^2=\frac{2K}{M'}\,, \qquad \varDelta^2=\frac{K}{M}\,,\\
&\omega_s{}^2=4\varDelta^2\sin^2\frac{\pi s}{2(N+1)}\,,\\
&A_s{}^2=\frac{1}{N+1}\,\omega_s{}^2\left(1-\frac{\omega_s{}^2}{4\varDelta^2}\right)=\frac{\varDelta^2}{N+1}\sin^2\frac{\pi s}{N+1}\,.
\end{aligned}\right\}\tag{5·3}$$

With the Hamiltonian brought into the form (5·2), the system can be considered as consisting of an isotopic atom coupled with $2N$ harmonic oscillators which are not coupled between each other. Under this form, the system was called *model S* and also studied by Toda and Kogure.[21] From the Hamiltonian (5·2), we deduce the canonical equations of motion

$$\dot{p}_s = -\omega_s^2 x_s - \omega_0 A_s x \qquad (s=-N, \cdots, -1, 1, \cdots N), \tag{5·4}$$

$$\dot{p} = -\omega_0^2 x - \omega_0 \sum_s A_s x_s\,. \tag{5·5}$$

What we shall essentially investigate concerning this dynamical system are the moment and continued fraction expansions of the Laplace transform of the classical autocorrelation function of the impurity momentum. This Laplace transform has for expression

$$\Xi(z) = \int_0^\infty \frac{(p(t), p)}{(p, p)} e^{-zt} dt\,. \tag{5·6}$$

The coefficients of the moment expansion and of the continued fraction expansion are given by

$$c_k = (\overset{k}{p}, \overset{k}{p})/(p, p)\,, \tag{5·7}$$

$$\left.\begin{aligned} &\Delta_k^2 = (f_k, f_k)/(f_{k-1}, f_{k-1})\,, \\ &\omega_k = 0\,, \end{aligned}\right\} \tag{5·8}$$

where $\overset{k}{p} = (d^k p(t)/dt^k)_{t=0}$, f_k is the k-th random force acting on p and the inner product is the classical canonical average. As $p(t)$ is Hermitian, all the coefficients ω_k vanish.

We shall start by calculating the random forces f_k. It has been recently pointed out by Sakurai[22] that $\Delta_1^2 = \omega_0^2$, $\Delta_2^2 = \Delta_3^2 = \Delta_4^2 = \Delta^2$ and, then, if one assumes that $\Delta_n^2 = \Delta^2$ for any $n > 1$, that the continued fraction expansion of (5·6) can be summed exactly. In the following we shall establish the expressions of the random forces to general order and show that indeed, in the limit $N \to \infty$ $\Delta_n^2 = \Delta^2$ for any $n > 1$.

In order to do so, we make use of the relations (3·8) and (3·9), to write down the expressions of the random forces of lowest order. The relations (3·8) and (3·9) appear to be here very practical, the sums $S_p^{(n)}$ being easy to calculate. The resulting expressions which involve sums of the form $\sum_s A_s^2 \omega_s^{2n}$ are

$$\left.\begin{aligned}
&f_1 = -\omega_0^2 x - \omega_0 \sum_s A_s x_s\,, && (f_1, f_1) = \omega_0^2/\beta\,, \\
&f_2 = -\omega_0 \sum_s A_s p_s\,, && (f_2, f_2) = \omega_0^2 \sum_s A_s^2/\beta\,, \\
&f_3 = \omega_0 \sum_s A_s \Big(\omega_s^2 - \sum_s A_s^2\Big) x_s, && (f_3, f_3) = \omega_0^2 \Big[\sum_s A_s^2 \omega_s^2 - \Big(\sum_s A_s^2\Big)^2\Big]/\beta, \\
&f_4 = \omega_0 \sum_s A_s \left(\omega_s^2 - \frac{\sum_s A_s^2 \omega_s^2}{\sum_s A_s^2}\right) p_s, && (f_4, f_4) = \omega_0^2 \left[\sum_s A_s^2 \omega_s^4 - \frac{(\sum_s A_s^2 \omega_s^2)^2}{\sum_s A_s^2}\right]/\beta,
\end{aligned}\right\} \tag{5·9}$$

where $\beta = (kT)^{-1}$, T being the absolute temperature and k Boltzmann's constant. The expressions of the random forces and their autocorrelation functions for higher orders become rather complicated, but in the limit $N\to\infty$, they simplify greatly because the sums $\sum_s \Lambda_s^2\omega_s^{2n}$ take the simple limiting value

$$\lim_{N\to\infty}\sum_s \Lambda_s^2\omega_s^{2n}=\frac{1}{n+2}\binom{2n+2}{n+1}\Delta^{2n+2}. \tag{5·10}$$

If we go over to this limit and then carefully inspect the resulting lowest order expressions—we went up to f_7 in our case—we are led to write for the general expressions in the same limit $N\to\infty$:

$$\left.\begin{aligned}
&f_1=-\omega_0^2x-\omega_0\sum_s \Lambda_s x_s,\\
&f_{2n-1}=(-1)^n\omega_0\sum_s \Lambda_s\Big[\omega_s^{2n-2}-\binom{2n-3}{1}\omega_s^{2n-4}\Delta^2+\cdots\\
&\quad+(-1)^p\binom{2n-2-p}{p}\omega_s^{2n-2-2p}\Delta^{2p}+\cdots+(-1)^{n-1}\Delta^{2n-2}\Big]x_s, \qquad (n>1),\\
&f_{2n}=(-1)^n\omega_0\sum_s \Lambda_s\Big[\omega_s^{2n-2}-\binom{2n-2}{1}\omega_s^{2n-4}\Delta^2+\cdots\\
&\quad+(-1)^p\binom{2n-1-p}{p}\omega_s^{2n-2-2p}\Delta^{2p}+\cdots+(-1)^{n-1}\binom{n}{n-1}\Delta^{2n-2}\Big]p_s,\ (n\geq 1),\\
&\Delta_1^2=\omega_0^2, \qquad \Delta_n^2=\Delta^2 \qquad (n>1).
\end{aligned}\right\} \tag{5·11}$$

with, for the sums $S_p^{(n)}$, the general expressions

$$S_p^{(n)}=\binom{n-p}{p-1}\omega_0^2\Delta^{2p-2}+\binom{n-p}{p}\Delta^{2p} \qquad (N\to\infty). \tag{5·12}$$

The proof of validity of the expressions (5·11) offers no difficulty: one just has to verify that they satisfy the recurrence equation (3·13), which is easily done.

Having calculated the random forces to general order, let us now turn to the moments. These also can be calculated to general order.

In the limit $N\to\infty$, this calculation can be done in two ways. A first way is to apply the formulae (3·17) and (3·18), since one knows the general expressions of the sums $S_p^{(n)}$, given by (5·12). This yields the following determinantal expressions for the even moments:

$$c_{2n}=\begin{vmatrix}
\omega_0^2\Delta^{4n-2}-[\omega_0^2+(2n-2)\Delta^2] & \cdots & (-1)^n\omega_0^2\Delta^{2n-2} & 0 & \cdots & 0 & 0\\
\omega_0^2\Delta^{4n-4} & 1\ \cdots & (-1)^{n-1}[(n-1)\omega_0^2+\Delta^2]\Delta^{2n-4} & 0 & \cdots & 0 & 0\\
\omega_0^2\Delta^{4n-6} & 0\ \cdots & (-1)^{n-2}\left[\frac{(n-1)(n-2)}{2}\omega_0^2+(n-1)\Delta^2\right]\Delta^{2n-6} & & & & \\
 & & & (-1)^{n-1}\omega_0^2\Delta^{2n-4} & \cdots & 0 & 0\\
 & & \cdots\cdots\cdots\cdots & & & & \\
\omega_0^2\Delta^2 & 0\ \cdots & 0 & 0 & \cdots & 1 & -\omega_0^2\\
\omega_0^2 & 0\ \cdots & 0 & 0 & & 0 & 1
\end{vmatrix} \tag{5·13}$$

and analogous expressions for the odd moments. The second way is to proceed directly, using repeatedly (5·4) and (5·5). After adequate rearrangements, one obtains

$$\left.\begin{aligned} (-1)^n \overset{2n}{p} &= A_n p + \omega_0 \sum_s A_s B_n^{(s)} p_s\,, \\ (-1)^n \overset{2n-1}{p} &= A_n x + \omega_0 \sum_s A_s B_n^{(s)} x_s\,, \end{aligned}\right\} \tag{5·14}$$

where A_n and B_n obey the recurrence relations

$$\left.\begin{aligned} &A_0=1, \qquad A_1=\omega_0^2, \\ &A_n=\omega_0^2 \Big(A_{n-1}+A_{n-2}\sum_s \varLambda_s^2+\cdots+A_{n-p}\sum_s \varLambda_s^2\omega_s^{2p-4}+\cdots+A_0\sum_s \varLambda_s^2\omega_s^{2n-4}\Big), \\ &\hspace{25em}(n\geq 2) \end{aligned}\right\} \tag{5·15}$$

$$\left.\begin{aligned} &B_0^{(s)}=0, \qquad B_1^{(s)}=1, \\ &B_n^{(s)}=\omega_0^2 \Big(B_{n-1}^{(s)}+B_{n-2}^{(s)}\sum_s \varLambda_s^2+\cdots+B_{n-p}^{(s)}\sum_s \varLambda_s^2\omega_s^{2p-4}+\cdots B_1^{(s)}\sum_s \varLambda_s^2\omega_s^{2n-6}\Big) \\ &\qquad +\omega_s^{2n-2}. \hspace{20em}(n\geq 2) \end{aligned}\right\} \tag{5·16}$$

Since from (5·14) one has

$$(-1)^n (\overset{2n}{p}, p) = (\overset{n}{p}, \overset{n}{p}) = A_n (p, p) \tag{5·17}$$

it follows from (5·15) that

$$c_n = \frac{(\overset{n}{p}, \overset{n}{p})}{(p, p)} = \begin{vmatrix} \omega_0^2 & \omega_0^2 \sum_s \varLambda_s^2 & \omega_0^2 \sum_s \varLambda_s^2\omega_s^2 & \cdots & \omega_0^2 \sum_s \varLambda_s^2\omega_s^{2n-4} \\ -1 & \omega_0^2 & \omega_0^2 \sum_s \varLambda_s^2 & \cdots & \omega_0^2 \sum_s \varLambda_s^2\omega_s^{2n-6} \\ 0 & -1 & \omega_0^2 & \cdots & \omega_0^2 \sum_s \varLambda_s^2\omega_s^{2n-8} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ 0 & 0 & 0 & \cdots & \omega_0^2 \end{vmatrix}. \tag{5·18}$$

We note that the expression (5·18) is valid for both finite and infinite values of N. If $N\to\infty$ the determinant (5·18) becomes

$$c_n = \begin{vmatrix} \omega_0^2 & \omega_0^2\varDelta^2 & 2\omega_0^2\varDelta^4 & \cdots & \frac{1}{n}\binom{2n-2}{n-1}\omega_0^2\varDelta^{2n-2} \\ -1 & \omega_0^2 & \omega_0^2\varDelta^2 & \cdots & \frac{1}{n-1}\binom{2n-4}{n-2}\omega_0^2\varDelta^{2n-4} \\ 0 & -1 & \omega_0^2 & \cdots & \frac{1}{n-2}\binom{2n-6}{n-3}\omega_0^2\varDelta^{2n-6} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ 0 & 0 & 0 & \cdots & \omega_0^2 \end{vmatrix}. \tag{5·19}$$

The fact that the determinants (5·13) and (5·19) have equal values is not easy to demonstrate generally, but can be easily verified for low values of n.

Now that we are in possession of both sets of quantities Δ_j^2 and c_k, we can investigate the convergence of the expansions of $\Xi(z)$. We find ourselves in the case where the coefficients Δ_j^2 are extremely simple, whereas the moment expressions are heavier to handle. Therefore we use the criteria given in terms of the quantities Δ_j^2.

Since $\lim_{n\to\infty} \Delta_n^2 = \Delta^2$, we conclude that there exists a fixed quantity l such that

$$\lim_{n\to\infty} \frac{(\overset{n}{p}, \overset{n}{p})}{(\underset{n-1}{p}, \underset{n-1}{p})} = l \tag{5·20}$$

and, using (3·27) and (4·8), we have

$$\omega_0^2 = \frac{c_1}{c_0} < \frac{c_n}{c_{n-1}} < l < 4\Delta^2 . \tag{5·21}$$

Therefore *the moment expansion does represent* $\Xi(z)$ *for* $|z| > \sqrt{l}$, Re $z>0$, *but is a divergent series for* $|z| \leq \sqrt{l}$.

For the convergence of the continued fraction, it follows from

$$k_{2p} = \frac{1}{\omega_0^2}\,, \qquad k_{2p+1} = \frac{\omega_0^2}{\Delta^2} \tag{5·22}$$

that the series $\sum_p k_p$ is divergent, that is, *the continued fraction expansion converges to* $\Xi(z)$ *for every* z *such that* Re $z>0$. To compute the function which it converges to, we use the well-known result[12)]

$$\cfrac{1}{1+\cfrac{(1+t)z}{1+\cfrac{z}{1+\cfrac{z}{1+\ddots}}}} = \frac{2}{\pi}\int_0^1 \frac{1+t}{(1+t)^2 - 4tu} \sqrt{\frac{1}{u}-1}\, \frac{du}{1+4zu} \tag{5·23}$$

to obtain

$$\Xi(z) = \frac{Q+1}{Qz + \sqrt{z^2+4\Delta^2}}\,, \tag{5·24}$$

$$\Xi_n(z) = \frac{(f_n(z), f_n)}{(f_n, f_n)} = \frac{1}{2\Delta^2}(\sqrt{z^2+4\Delta^2} - z), \qquad (n \geq 1), \tag{5·25}$$

where we have taken for the radical the positive determination: as pointed out in § 4, we find indeed branch points on the imaginary axis.

So far, we have not made explicit mention of the fact that the isotopic impurity had to be heavy in order to exhibit a Brownian motion. Let us then consider the limiting case where $Q\to\infty$, $\Delta\to\infty$ in such a way that[18)]

$$\lim_{\substack{Q\to\infty \\ \Delta\to\infty}} \frac{2\Delta}{1+Q} = \lim_{\substack{Q\to\infty \\ \Delta\to\infty}} \frac{\omega_0^2}{\Delta} = \text{const}\,. \tag{5·26}$$

As shown by (5·21), the lower bound of l goes to infinity like Δ, whereas the upper bound goes also to infinity, but much faster, like Δ^2:*) *that is the moment expansion becomes asymptotic. However*, as shown by (5·22), the series $\sum_p k_p$ remains divergent and *the continued fraction expansion remains convergent. Thus we have clearly demonstrated on this particular example, how the continued fraction representation can play the role of an analytic continuation for the moment expansion.*

Before closing this section, we would like to investigate briefly the analytic behaviour of $\Xi(z)$ in the half-plane $\mathrm{Re}\, z<0$, especially with respect to Mori's long-time approximation. We shall mainly do it in the frame of the limiting case (5·26), for which ω_0^2/Δ^2 is an infinitesimally small quantity.

First of all we see that if we continue the function (5·24) into the half plane $\mathrm{Re}\, z<0$ on the Riemann sheet corresponding to the plus sign determination for the radical in the denominator, $\Xi(z)$ has a pole exactly given by

$$z_0 = -\frac{\omega_0^2}{\Delta}\left(1-\frac{\omega_0^2}{\Delta^2}\right)^{-1/2} = -\frac{\omega_0^2}{\Delta} - \frac{\omega_0^4}{2\Delta^3} + O\left(\frac{1}{\Delta^2}\right). \tag{5·27}$$

Now, to introduce Mori's n-th long time approximation means that writing $\Xi(z)$ as

$$\Xi(z) = \cfrac{1}{z+\cfrac{\Delta_1^2}{z+\ddots+\cfrac{\Delta_{n-1}^2}{z+\Xi_n(z)}}}\,, \tag{5·28}$$

where $\Xi_n(z)$ is defined by Eq. (5·25), we neglect the z dependence of $\Xi_n(z)$ and take

$$\Xi_n(z) \approx \Xi_n(0)\,. \tag{5·29}$$

For the present model, we deduce from (5·25):

$$\Xi_1(0) = \Xi_2(0) = \cdots = \Xi_n(0) = \cdots = \Delta^{-1}\,. \tag{5·30}$$

Thus the first approximation yields

$$\Xi^{(1)}(z) = \frac{1}{z+\omega_0^2/\Delta} \tag{5·31}$$

to which corresponds a pole at

*) Actually it can be seen from the expression (5·19) that

$$\lim_{\substack{Q\to\infty \\ \Delta\to\infty}} \lim_{\omega_0^2/\Delta=\text{const}} \lim_{n\to\infty} \frac{1}{4\Delta^2}\frac{c_n}{c_{n-1}} = 1\,.$$

$$z_0^{(1)} = -\omega_0^2/\Delta\,. \tag{5·32}$$

The second approximation yields

$$\Xi_2(z) = \frac{z+\Delta}{z^2+z\Delta+\omega_0^2} \tag{5·33}$$

with poles at

$$z_1^{(2)} = -\frac{\omega_0^2}{\Delta} - \frac{\omega_0^4}{\Delta^3} + O\left(\frac{1}{\Delta^2}\right), \qquad z_2^{(2)} = -\Delta + \frac{\omega_0^2}{\Delta} + O\left(\frac{1}{\Delta}\right). \tag{5·34}$$

Finally the third approximation is

$$\Xi^{(3)}(z) = \frac{z^2+z\Delta+\Delta^2}{z^3+z^2\Delta+z(\omega_0^2+\Delta^2)+\omega_0^2\Delta} \tag{5·35}$$

with poles at

$$\left.\begin{aligned} &\left.\begin{matrix} z_1^{(3)} \\ z_2^{(3)} \end{matrix}\right\} = -\frac{\Delta}{2} + \frac{\omega_0^2}{2\Delta} + O\left(\frac{1}{\Delta^2}\right) \pm \frac{i\sqrt{3}}{2}\left[\Delta + \frac{\omega_0^2}{3\Delta} + O\left(\frac{1}{\Delta}\right)\right], \\ &z_3^{(3)} = -\frac{\omega_0^2}{\Delta} + O\left(\frac{1}{\Delta^2}\right). \end{aligned}\right\} \tag{5·36}$$

We see that in all the first three approximations, we obtain one pole near the origin and close to the exact pole and other poles very far away: this result confirms perfectly Mori's general predictions.[2] Namely, introducing the sequence of constants defined by

$$\Xi(0) = \frac{1}{\lambda_0}, \qquad \lambda_{j-1} = \frac{\Delta_j^2}{\lambda_j}\,(j \geq 1) \tag{5·37}$$

we should expect, if $\lambda_n \gg \lambda_{n-1}, \cdots, \lambda_1, \lambda_0$, n poles located near the origin. In the present case, we have

$$\lambda_0 = \omega_0^2/\Delta\,, \qquad \lambda_1 = \lambda_2 = \cdots = \lambda_n = \cdots = \Delta \tag{5·38}$$

and we find in the limiting case (5·26), for which $\lambda_1 \gg \lambda_0$, only one pole located near the origin. As for the other poles far away from the origin appearing in the second and third order approximations, they do not correspond to any singularity of $\Xi(z)$: the possible existence of such meaningless poles was also suggested by Mori.[2]

§ 6. On the general convergence properties of the representations of time autocorrelation functions

The example treated in the previous section has shown us that the moment expansion could have a finite radius of convergence or could be an asymptotic series, whereas in both cases the continued fraction expansion was representing the relaxation function in the entire right half-plane.

A first question which arises is the following: In case of convergence, which expansion does converge faster?

We shall restrict ourselves to the case of Hermitian variables. Thus we can use inequalities established by Stieltjes[14] in the case of a real variable x. Applying them to our problem, we obtain

$$\frac{c_0}{x}-\frac{c_1}{x^3}+\cdots-\frac{c_{2n-1}}{x^{4n-1}}<\frac{N_{2n}(x)}{M_{2n}(x)}<\Xi(x)<\frac{N_{2n+1}(x)}{M_{2n+1}(x)}<\frac{c_0}{x}-\frac{c_1}{x^3}+\cdots+\frac{c_{2n}}{x^{4n+1}}, \tag{6·1}$$

where $M_n(x)$ and $N_n(x)$ are the denominator and numerator of the n-th approximant of the continued fraction, as defined by (2·15). It is always advantageous to use the continued fraction to approximate $\Xi(x)$, because the successive approximants give closer values than the successive sums of the series. We may note that, even if the moment expansion is divergent, one can use it to obtain approximations of $\Xi(x)$: sums of an even number of terms will give us lower values, whereas sums with an odd number of terms will give us higher values. The same holds for the approximants of the continued fraction.

A second question that one can ask is the following: is it possible that for any dynamical variable and all systems usually encountered in Statistical Mechanics, the continued fraction expansion would be convergent for any z such that $\mathrm{Re}\, z>0$? If the answer is yes, then, beyond the convergence problem, we shall have obtained a new general property of time autocorrelation functions, this property being best expressed through the condition imposed upon the moments. In this way we shall have attained in one point the ultimate goal of our convergence study, namely to discover new elements of information on the general behaviour of time autocorrelation functions.

For this second question, we shall restrict ourselves again to the case of Hermitian variables. Before trying to answer the question, let us first discuss some implications and consequences of an affirmative answer.

An affirmative answer implies that the series $\sum_n c_n^{-1/2n}$ be divergent for any Hermitian dynamical variable attached to any statistical mechanical system. It is well known[23] in the theory of Probability that the quantity $c_n^{1/2n}$ increases with n: this is readily proved by using Hölder's inequality. Thus, for the series $\sum_n c_n^{-1/2n}$ to be divergent, the quantity $c_n^{1/2n}$ must not increase too rapidly. This amounts to say that $\xi(\omega)$ must not be too spread out, or equivalently that $\Xi(t)$ cannot decrease too steeply near the initial time.

Now it is not easy to deduce from this divergence condition quantitative informations on the general behaviour of $\Xi(t)$ or $\xi(\omega)$. The reason simply is that all the known criteria of convergence or divergence yield sufficient conditions but not necessary conditions.

It is $\xi(\omega)$ which seems most suitable for some quantitative statements: this is to be expected since it is the distribution function determining the moments c_n. Indeed, as it is customary to do when studying the rate of growth of a function, let us take the exponential functions $\exp(-a|\omega|^{\alpha})$ $(a>0)$ for reference functions: then it is known[24] that the series $\sum_n c_n^{-1/2n}$ is divergent if and only if $\alpha\geq 1$. If the distribution is more widely spread out $(\alpha<1)$, then one can add to $\xi(\omega)$ a multiple of a function such that $\exp(-|\omega|^{1/2})\cos|\omega|^{1/2}$, all the moments of which are zero, so that the total moments are still the same: the moment problem thus is not determinate any more, the series $\sum_n c_n^{-1/2n}$ no more divergent and the continued fraction no more convergent.

There is a case where the previous statements can be verified and quantitatively discussed. It is the case of a *Gaussian relaxatsion* for which both $\Xi(t)$ and $\xi(\omega)$ are Gaussian. If

$$\Xi(t)=\exp(-\Delta^2 t^2/2)\ , \tag{6·2}$$

then one finds that

$$c_n=(1.\ 3.\ 5\cdots 2n-1)\Delta^{2n} \tag{6·3}$$

and the series $\sum_n c_n^{-1/2n}$ diverges. Thus the continued fraction expansion converges, whereas the moment expansion is asymptotic, since $c_{n+1}/c_n=(2n+1)\Delta^2$. Moreover, the coefficients Δ_n^2 have simple expressions

$$\Delta_n^2=n\Delta_1^2=n\Delta^2 \tag{6·4}$$

which illustrate the result of Stieltjes, according to which c_{n+1}/c_n has no upper limit if Δ_n^2 increases without bound with n.

Another point we would like to mention is the connection between the convergence of the moment expansion of the relaxation function itself and the convergence of the continued fraction expansion of its Laplace transform. Indeed, *if the series* (1·13) *is analytic near the origin, then the continued fraction expansion is convergent for z such that* $\mathrm{Re}\, z>0$. The proof is simple. If the series (1·13) is analytic near the origin, then[26] it is sufficient and necessary that there exist two numbers M and δ such that for any n

$$|c_n|<M\delta^{2n}(2n)!\,. \tag{6·5}$$

This inequality has for consequence that the series $\sum_n c_n^{-1/2n}$ is divergent and therefore the continued fraction converges for z such that $\mathrm{Re}\, z>0$.

These remarks on the implications of the convergence of the continued fraction expansion being made, let us come to the question of the proof.

In order to study the convergence of the series $\sum_n c_n^{-1/2n}$, we have chosen *for inner product Kubo's canonical product*, as defined by (4·4) and tried to find upper bounds for the moments c_n in the case of *an Hermitian quantum dynamical variable* $A(t)$. Indeed, it may be very difficult to calculate a quantity but much easier to find an upper bound for it. The details of the demonstrations are given in the Appendix and in the following we shall only state and discuss the results obtained.

The expression of $(\overset{n}{A}, \overset{n}{A})$ can be brought into the form

$$(\overset{n}{A}, \overset{n}{A}) = \frac{1}{\beta}\int_0^\beta \langle e^{\lambda H}\overset{n}{A} e^{-\lambda H}\overset{n}{A}\rangle d\lambda = \frac{2}{\beta\hbar^{2n}}\Big[\langle AH^{2n-1}A\rangle - \binom{2n-1}{1}\langle HAH^{2n-2}A\rangle + \cdots$$

$$+ (-1)^p \binom{2n-1}{p}\langle H^p AH^{2n-1-p}A\rangle - \langle H^{2n-1}AA\rangle\Big]. \qquad (6\cdot6)$$

The crudest way of finding an upper bound for this expression is to replace every term by the largest with a positive sign. This leads to the exact inequality

$$c_n \leq \frac{M}{\beta}\left(\frac{2}{\hbar}\right)^{2n} \langle H^{4n-2}\rangle^{1/2}, \qquad (6\cdot7)$$

where M is a dimensionless constant, independent of n.

To go further, we must calculate the n-dependence of $\langle H^{4n-2}\rangle$. Assuming the system large enough so that we can treat the energy spectrum as continuous, we have

$$\langle H^{4n-2}\rangle = \int_0^\infty E^{4n-2} e^{-\beta E}\rho(E)\,dE, \qquad (6\cdot8)$$

where $\rho(E)$ is the number of eigenstates with energy between E and $E+dE$. For an exact estimation of the right-hand side of the inequality $(6\cdot7)$, it becomes at this point necessary to specify the system in order to know $\rho(E)$.

For simplicity let us first consider the case of a perfect gas of N particles of mass m in a volume V and let us assume that we are in conditions such that we can take for $\rho(E)$ the quasi-classical value

$$\rho(E) = \frac{3N}{2}\frac{V^N}{N!}\left(\frac{2\pi m}{h^2}\right)^{3N/2}\frac{E^{3N/2-1}}{\Gamma(3N/2+1)}. \qquad (6\cdot9)$$

If keeping N fixed, we let n become very large, then we find that

$$c_n^{-1/2n} \geq \frac{\beta\hbar}{4}\frac{1}{2n-1}. \qquad (6\cdot10)$$

Therefore for a classical perfect gas of a finite number of particles, the series $\sum_n c_n^{-1/2n}$ is divergent. The same conclusion holds for a finite system of weakly interacting harmonic oscillators of frequency ν, for which[27]

$$\rho(E) = N\left(\frac{1}{h\nu}\right)^N \frac{1}{\Gamma(N+1)} E^{N-1}. \qquad (6\cdot11)$$

More generally, for systems usually encountered in Statistical Mechanics, it has been suggested[27] or assumed[28] that the increase of $\rho(E)$ with energy can be represented by a function of the form

$$\rho(E) = CE^{N'}, \tag{6·12}$$

where N' is a fixed number of the order of the number of degrees of freedom and C a constant. Now, for such an energy dependence of $\rho(E)$, the series $\sum_n c_n^{-1/2n}$ is divergent. Thus, *if the assumption* (6·12) *is correct, the convergence of the continued fraction expansion in the right half-plane would hold for any Hermitian dynamical variable attached to a usual statistical mechanical system of finite size.*

We can still state our result in the following way: *For any Hermitian dynamical variable attached to a system of perhaps very large but finite size which obeys* (6·12), *the continued fraction expansion of the Laplace transform of the relaxation function is convergent for any z such that* $\mathrm{Re}\, z > 0$.

In the limiting case where $N \to \infty$, $V \to \infty$, $V/N = \text{const}$, the inequality (6·7) leads to the trivial result $c_n^{-1/2n} \geq 0$. The reason is that the expression (6·6) has been treated in a too simple way. Indeed this expression can be rewritten as

$$\begin{aligned}(\overset{n}{A}, \overset{n}{A}) = \frac{2}{\beta\hbar^{2n}}\Big[&\langle AH^{2n-1}A - AAH^{2n-1}\rangle - \binom{2n-1}{1}\langle HAH^{2n-2}A - AHAH^{2n-2}\rangle \\ &+ \cdots\cdots\cdots \\ &+ (-1)^p \binom{2n-1}{p}\langle H^p AH^{2n-1-p}A - AH^p AH^{2n-1-p}\rangle \\ &+ \cdots\cdots\cdots \\ &+ (-1)^{n-1}\binom{2n-1}{n-1}\langle H^{n-1}AH^n A - AH^{n-1}AH^n\rangle \Big].\end{aligned} \tag{6·13}$$

Looking for the largest of the differences appearing in the right-hand side, we obtain the inequality

$$c_n \leq \frac{1}{2\beta}\left(\frac{2}{\hbar}\right)^{2n} \frac{\langle A^2\rangle^{1/2}}{(A, A)} \langle H^{2m}[A, H^{2n-1-m}]^2\rangle^{1/2}, \tag{6·14}$$

where $m < 2n$ may be zero or not. Now the upper limit found for c_n depends upon the average of a quantity which does not increase to infinity when the system becomes infinitely large, because it contains the commutator of A with H^{2n-1} or H^{2n-1-m} and is not simply H^{4n-2}.

If the expression (6·6) has then be treated in a more adequate way, we are now facing a much more difficult problem in trying to evaluate the n-dependence of the upper bound (6·14). This problem is being presently investigated. Although we cannot yet offer a rigorous justification, we think it likely that under the assumption (6·12), the continued fraction expansion remains convergent, if not for any Hermitian dynamical variable, at least for very general classes of Hermitian variables, even if the size of the system becomes infinitely large.

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Appendix

The problem is to find an upper bound for the moment $c_n = (\overset{n}{A}, \overset{n}{A})/(A, A)$, where the inner product is Kubo's canonical product (4·4).

Using repeatedly Heisenberg's equation of motion, we have

$$(i\hbar)^{2n}\overset{2n}{A} = [\cdots[[[A, H], H], H]\cdots] =$$
$$AH^{2n} - \binom{2n}{1}HAH^{2n-1} + \binom{2n}{2}H^2AH^{2n-2} - \cdots + H^{2n}A\,. \qquad \text{(A·1)}$$

Let us then define the function

$$A(\lambda) = e^{\lambda H} A\, e^{-\lambda H} \qquad \text{(A·2)}$$

the $2n$-th derivative of which has for expression

$$A^{(2n)}(\lambda) = H^{2n}e^{\lambda H}Ae^{-\lambda H} - \binom{2n}{1}H^{2n-1}e^{\lambda H}AHe^{-\lambda H}$$
$$+ \binom{2n}{2}H^{2n-2}e^{\lambda H}AH^2e^{-\lambda H} + \cdots + e^{\lambda H}AH^{2n}e^{-\lambda H}\,. \qquad \text{(A·3)}$$

As expected, $(i\hbar)^{2n}\overset{2n}{A} = A^{(2n)}(0)$, since $A(it/\hbar)$ is the formal solution of Heisenberg's equation. Using (A·1) and (A·3), we may write

$$\hbar^{2n}(\overset{n}{A}, \overset{n}{A}) = (i\hbar)^{2n}(\overset{2n}{A}, A) = \frac{(i\hbar)^{2n}}{\beta}\int_0^\beta \langle e^{\lambda H}\overset{2n}{A}\, e^{-\lambda H}A^\dagger\rangle d\lambda$$
$$= \frac{1}{\beta}\int_0^\beta \langle A^{(2n)}(\lambda)\, A^\dagger\rangle d\lambda = \frac{1}{\beta}\Big[\langle A^{(2n-1)}(\beta)\, A^\dagger\rangle - \langle A^{(2n-1)}(0)\, A^\dagger\rangle\Big]. \qquad \text{(A·4)}$$

Introducing the partition function $Z = \mathrm{Tr}\, e^{-\beta H}$, Eq. (A·4) can be written:

$$\beta Z\hbar^{2n}(\overset{n}{A}, \overset{n}{A}) = \Big[\mathrm{Tr}(H^{2n-1}Ae^{-\beta H}A^\dagger) - \binom{2n-1}{1}\mathrm{Tr}(H^{2n-2}AHe^{-\beta H}A^\dagger)$$
$$+ \cdots - \mathrm{Tr}(AH^{2n-1}e^{-\beta H}A^\dagger)\Big]$$
$$- \Big[\mathrm{Tr}(H^{2n-1}e^{-\beta H}AA^\dagger) - \binom{2n-1}{1}\mathrm{Tr}(H^{2n-2}e^{-\beta H}AHA^\dagger)$$
$$+ \cdots - \mathrm{Tr}(e^{-\beta H}AH^{2n-1}A^\dagger)\Big] \qquad \text{(A·5)}$$

or, since the trace of a product is invariant under cyclic permutation of the factors,

$$\beta Z\hbar^{2n}(\overset{n}{A},\overset{n}{A})=\mathrm{Tr}\,[H^{2n-1}(Ae^{-\beta H}A^\dagger+A^\dagger e^{-\beta H}A)]$$
$$-\binom{2n-1}{1}\mathrm{Tr}[H^{2n-2}(Ae^{-\beta H}HA^\dagger+A^\dagger e^{-\beta H}HA)]$$
$$+\cdots\cdots\cdot$$
$$-\mathrm{Tr}[Ae^{-\beta H}H^{2n-1}A^\dagger+A^\dagger e^{-\beta H}H^{2n-1}A]\,. \tag{A·6}$$

From now on, we restrict ourselves to Hermitian variables; thus (A·6) reduces to

$$\tfrac{1}{2}\beta Z\hbar^{2n}(\overset{n}{A},\overset{n}{A})=\mathrm{Tr}\,(H^{2n-1}Ae^{-\beta H}A)$$
$$-\binom{2n-1}{1}\mathrm{Tr}\,(H^{2n-2}Ae^{-\beta H}HA)+\cdots-\mathrm{Tr}\,(Ae^{-\beta H}H^{2n-1}A)\,. \tag{A·7}$$

Furthermore, we shall assume that the energy of the system is defined in such a way that all the eigenvalues of the Hamiltonian are positive. It follows that all the terms of the right-hand side of (A·7) are positive. Hence, if we want to find an upper bound for $(\overset{n}{A},\overset{n}{A})$, we must find which is the largest term. Defining x_p by

$$x_p=\mathrm{Tr}\,(H^{2n-1-p}AH^pe^{-\beta H}A)\,, \tag{A·8}$$

we first show that the sequence of quantities x_p is convex, that is,

$$x_p{}^2\leq x_{p-1}x_{p+1}\,. \tag{A·9}$$

In order to prove it, we make use of a well-known inequality in the theory of linear operators[25] and write

$$[\mathrm{Tr}\,(H^{2q+1}AH^{2p+1}e^{-\beta H}A)]^2\equiv[\mathrm{Tr}\,(H^qAH^{p+1}e^{-\beta H/2}e^{-\beta H/2}H^pAH^{q+1})]^2$$
$$\leq\mathrm{Tr}\,(H^qAH^{p+1}e^{-\beta H}H^{p+1}AH^q)\,\mathrm{Tr}\,(H^{q+1}AH^pe^{-\beta H}H^pAH^{q+1})\,, \tag{A·10}$$

whence, by choosing p and q such that $p+q=2n-1$,

$$\frac{\mathrm{Tr}\,(H^{2q}AH^{2p+2}e^{-\beta H}A)}{\mathrm{Tr}\,(H^{2q+1}AH^{2p+1}e^{-\beta H}A)}\geq\frac{\mathrm{Tr}\,(H^{2q+1}AH^{2p+1}e^{-\beta H}A)}{\mathrm{Tr}\,(H^{2q+2}AH^{2p}e^{-\beta H}A)}\geq\cdots$$
$$\geq\frac{\mathrm{Tr}\,(H^{2n-2}AHe^{-\beta H}A)}{\mathrm{Tr}\,(H^{2n-1}Ae^{-\beta H}A)}=\frac{x_1}{x_0} \tag{A·11}$$

which establishes (A·9). This inequality means that x_p cannot go through a maximum: it may decrease steadily as p increases, or increase steadily or go through a minimum.

If the sequence x_p decreases steadily, the largest term is x_0 and we may write

$$\tfrac{1}{2}\beta Z\hbar^{2n}(\overset{n}{A},\overset{n}{A})\leq\left(1+\sum_{p=1}^{2n-1}\binom{2n-1}{p}\right)\mathrm{Tr}\;H^{2n-1}Ae^{-\beta H}A \tag{A·12}$$

or

$$\beta\hbar^{2n}(\overset{n}{A},\overset{n}{A})\le 2^{2n}\langle H^{4n-2}\rangle^{1/2}\langle Ae^{-\beta H}Ae^{\beta H}Ae^{-\beta H}A\rangle^{1/2}. \tag{A·13}$$

If the sequence x_p increases steadily, or goes through a minimum, then one can always choose n large enough so that x_n is the largest term. Thus one has

$$\tfrac{1}{2}\beta Z\hbar^{2n}(\overset{n}{A},\overset{n}{A})\le\left(1+\sum_{p=1}^{2n-1}\binom{2n-1}{p}\right)\mathrm{Tr}\,AH^{2n-1}e^{-\beta H}A \tag{A·14}$$

or

$$\beta\hbar^{2n}(\overset{n}{A},\overset{n}{A})\le 2^{2n}\langle H^{4n-2}\rangle^{1/2}\langle A^4\rangle^{1/2}. \tag{A·15}$$

In both cases (A·13) and (A·15), we thus find that

$$c_n\le\frac{M}{\beta}\left(\frac{2}{\hbar}\right)^{2n}\langle H^{4n-2}\rangle^{1/2}, \tag{A·16}$$

where M is a dimensionless constant, independent of n.

However, coming back to the expression (A·7), we note that we can write it

$$\begin{aligned}\tfrac{1}{2}\beta Z\hbar^{2n}(\overset{n}{A},\overset{n}{A}) &= \mathrm{Tr}\,(H^{2n-1}Ae^{-\beta H}A-AH^{2n-1}e^{-\beta H}A)\\ &\quad-\binom{2n-1}{1}\mathrm{Tr}\,(H^{2n-2}AHe^{-\beta H}A-HAH^{2n-2}e^{-\beta H}A)\\ &\quad+\cdots\cdots\cdots\\ &\quad+(-1)^{n-1}\binom{2n-1}{n-1}\mathrm{Tr}\,(H^nAH^{n-1}e^{-\beta H}A-H^{n-1}AH^ne^{-\beta H}A).\end{aligned} \tag{A·17}$$

If the sequence is steadily increasing or decreasing, one may write

$$\tfrac{1}{2}\beta Z\hbar^{2n}(\overset{n}{A},\overset{n}{A})\le\left(1+\sum_{p=1}^{n-1}\binom{2n-1}{p}\right)|\mathrm{Tr}\,(H^{2n-1}Ae^{-\beta H}A-AH^{2n-1}e^{-\beta H}A)|, \tag{A·18}$$

or

$$(\overset{n}{A},\overset{n}{A})\le\frac{1}{\beta}\frac{2^{2n-1}}{\hbar^{2n}}|\langle A[H^{2n-1},A]\rangle|, \tag{A·19}$$

whereas if the sequence is going through a minimum value equal to x_m, one may find n large enough so that

$$\tfrac{1}{2}\beta Z\hbar^{2n}(\overset{n}{A},\overset{n}{A})\le\left(1+\sum_{p=1}^{n-1}\binom{2n-1}{p}\right)\mathrm{Tr}\,(AH^{2n-1}e^{-\beta H}A-H^{2n-1-m}AH^me^{-\beta H}A) \tag{A·20}$$

or

$$(\overset{n}{A},\overset{n}{A})\le\frac{1}{\beta}\frac{2^{2n-1}}{\hbar^{2n}}\langle A[A,H^{2n-1-m}]H^m\rangle. \tag{A·21}$$

In both cases (A·19) and (A·21), we find that

$$c_n \leq \frac{1}{2\beta}\left(\frac{2}{\hbar}\right)^{2n} \frac{\langle A^2 \rangle^{1/2}}{(A, A)} \langle H^{2m}[A, H^{2n-1-m}]^2 \rangle^{1/2}, \tag{A·22}$$

where $m<2n$ may be zero or not.

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