PHYSICAL REVIEW B VOLUME 15, NUMBER 11 1 JUNE 1977

New method for calculating transport coefficients and application to the Heisenberg chain

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(Received 14 February 1977)

In this article, a new simple method to calculate memory functions within the Mori formalism is presented. It is then used to derive the relaxation function for a Heisenberg chain. The results for different choices of relevant variables are compared with experiments and other theories. It is found that for the right choice of variables, the results are in much better agreement with recent experiments on tetramethylammonium-manganese-trichloride (TMMC) than the results of other theories.

I. INTRODUCTION

During the last five years a lot of work has been devoted to the study of dynamical properties of one-dimensional spin systems. In these systems no spontaneous magnetization at nonzero temperatures exists[1] and consequently the simple spin-wave theory[2] cannot predict spin waves in contradiction with a lot of experiments.[3]

From the theoretical point of view the situation is not clear. For one-dimensional Heisenberg antiferromagnets, such as tetramethylammonium-manganese-trichloride (TMMC) and $\mathrm{CuCl_2 \cdot 2N(C_5D_5)}$, continued-fraction expansions[4,5] have been used to describe the dynamical properties. The static quantities which determine the continued-fraction expansion have been calculated exactly[5,6] for the classical one-dimensional Heisenberg system. At the moment, there are no similar results for planar systems.[7] In Ref. 4 the authors introduced a termination function in order to obtain a three-pole approximation. Some authors[5] prefer a Gaussian assumption, but in both cases they fail to predict spin waves for certain q values and certain temperatures, in disagreement with recent neutron-scattering experiments.[8] Another way of calculating relaxation functions is given by the mode-mode coupling equations.[9] However, one should realize that these equations are probably the simplest equations one can derive from Mori's formalism[10] after making some approximations.[11] In addition, a lot of phenomenologic assumptions are needed and although the mode-mode coupling scheme is systematic itself, a calculation of the relaxation function in successive approximations is impossible without making questionable suppositions.

On the other hand, numerical results for classical systems are known from computer calculations[12] and it follows that there is a serious disagreement between the theoretical values and the value obtained by computer simulation for the diffusion coefficient at infinite temperature.

Except for the direct computer calculations, we feel that the methods mentioned above fail because one has to make a crucial approximation for the transport coefficients. From Ref. 10 it is clear that the time evolution that determines the transport coefficients is not the same as the time evolution of the total system. Therefore the main purpose of this work is to show how one can evaluate these quantities with the correct time evolution in a simple and systematic way using the Mori formalism as a starting point.

In Sec. II, we recall some well-known results of the Mori formalism and we give a rather abstract but very general derivation of the equations from which the frequency-dependent transport coefficients can be calculated. This general formulation makes it much easier to apply this scheme for other physical systems. We show that if one makes only one reasonable approximation, the transport coefficients are uniquely determined by the moments of the relaxation functions. In Sec. III we write the Hamiltonian for the one-dimensional Heisenberg magnet and we give the relevant variables. In Sec. IV we give the dynamic equations and we explicitly show how the general theory works in this particular example. We make a detailed comparison with the experimental data for TMMC using the classical results for the moments. Finally the fluctuation-dissipation theorem is used to calculate the static quantities for the quantum-mechanical system.

II. FORMALISM

A. General theory

Let F denote the Hilbert space of operators A_μ; $\mu = 1, \ldots$ with scalar product (;). The time evolution in F is determined by the Hermitian Liouville operator L which is related to the Hamiltonian of the system by $LA=[H,A]$ or equivalently by $A(t)=e^{iLt}A(0)=e^{iHt}A(0)e^{-iHt}$. If we are only interested in the motion of some operators A_i, $i=1,\ldots,n$ the projector on this n-dimensional subspace E is given by[10]

$$PB=A_j X_{ji}^{-1}(A_i;B)\,,\quad B\in F\,. \tag{2.1}$$

Here we used the sum convention and we denoted X_{ij}^{-1} for the inverse of the matrix $(A_i;A_j)$; $i,j=1,\ldots,n$. If Q is the projector on the orthogonal complement of E we have the following properties

$$Q=1-P\,,\quad P=P^{\dagger}=P^2\,,\quad Q=Q^{\dagger}=Q^2\,. \tag{2.2}$$

If we want to know how the resolvent of L, given by $(z-L)^{-1}$, $\mathrm{Im}z>0$, which contains all information about the system, acts on the subspace E we can use the trivial identity $z(z-L)^{-1}=1+L(z-L)^{-1}$ and the definition of the projectors in order to obtain the set of coupled equations[13]

$$(zP-PLP)(z-L)^{-1}P-PLQ\,Q(z-L)^{-1}P=P\,; \tag{2.3a}$$

$$(zQ-QLQ)Q(z-L)^{-1}P=QLP\,P(z-L)^{-1}P\,. \tag{2.3b}$$

Substituting the expression for $Q(z-L)^{-1}P$ obtained from Eq. (2.3b) into Eq. (2.3a) we find

$$[zP-PLP-PLQ(z-QLQ)^{-1}QLP]P(z-L)^{-1}P=P\,. \tag{2.4}$$

Taking the i,j matrix element we have

$$[z\delta_{ik}-(A_i;LA_l)X_{lk}^{-1}-(QLA_i;(z-QLQ)^{-1}QLA_l)X_{lk}^{-1}]$$
$$\times(A_k;(z-L)^{-1}A_j)=(A_i;A_j)\,, \tag{2.5}$$

where $i,j,k,l=1,\ldots,n$.

Although it is not necessary,[10] one can define the scalar product as the static susceptibility $\chi_{A_iA_j}(0)$, and then $(A_k;(z-L)^{-1}A_j)$ represents the relaxation function, while $(A_i;LA_l)X_{lk}^{-1}$ and $(QLA_i;(z-QLQ)^{-1}QLA_l)X_{lk}^{-1}$ correspond to the frequency matrix and memory matrix, respectively. Now we have to evaluate the memory matrix. In principle, this is as difficult as a direct calculation of the relaxation function itself. Because the time evolution has been separated into a dominant part (the frequency matrix) and a perturbation (the memory matrix) one might expect that it is easier to find an approximation for the memory matrix than to approximate the relaxation function itself.[10,13] However, there is an additional difficulty because the time evolution of the memory matrix is determined by QLQ. If it is possible to use perturbation theory it is sometimes allowed to omit Q operators for small wave vectors,[13] but such an approximation leads to unreasonable results for spin systems in the paramagnetic region.[11] Therefore we present a simple method which takes a chosen number of Q's into account.

Defining the function $f_{ij}(z)$ by

$$f_{ij}(z)=(QLA_i;(z-QLQ)^{-1}QLA_j)\,, \tag{2.6}$$

and using the identity $z(z-QLQ)^{-1}=1+QLQ(z-QLQ)^{-1}$ twice, we easily obtain

$$(z^2\delta_{ik}+z\Omega_{ik}^{(1)}+\Omega_{ik}^{(2)})f_{kj}(z)$$
$$=z(QLA_i;QLA_j)+(QL^2A_i;QLA_j)$$
$$+(QL^3A_i;(z-QLQ)^{-1}QLA_j)\,, \tag{2.7}$$

where we denoted $\Omega_{ik}^{(n)}=(A_i;L^nA_l)X_{lk}^{-1}$.

In principle we can continue in this way, but since we have to stop anyhow, we need an approximation. The simplest thing we can do is to replace the third term on the right-hand side of Eq. (2.7) by a complex number R_{ij} and we have

$$(z^2\delta_{ik}+z\Omega_{ik}^{(1)}+\Omega_{ik}^{(2)})f_{kj}(z)$$
$$=z(QLA_i;LA_j)+(QL^2A_i;LA_j)+R_{ij}\,. \tag{2.8}$$

Taking the limit $z=i\epsilon$, $\epsilon\to0$ we find

$$\Omega_{ik}^{(2)}f_{kj}(i\epsilon)=(QL^2A_i,LA_j)+R_{ij}\,. \tag{2.9}$$

Substituting this equation for R_{ij} into Eq. (2.8) yields

$$(z^2\delta_{ik}+z\Omega_{ik}^{(1)}+\Omega_{ik}^{(2)})f_{kj}(z)=z(QLA_i;LA_j)+\Omega_{ik}^{(2)}f_{kj}(i\epsilon)\,. \tag{2.10}$$

Since QLQ is a Hermitian operator we can use Eq. (B3) of Appendix B immediately for $\varphi(\omega)=1$ and consequently we can write

$$(QLA_i;LA_j)$$
$$=\frac{-1}{2\pi i}\lim_{\epsilon\to0}\int_{-\infty}^{+\infty}d\omega\,[f_{ij}(\omega+i\epsilon)-f_{ij}(\omega-i\epsilon)]\,. \tag{2.11}$$

Now it is clear that Eqs. (2.10) and (2.11) constitute a linear set of coupled equations for $f_{ij}(i\epsilon)$. For simple systems such as phonon or spin systems these equations can be solved analytically as will be shown later on.

If we want to calculate the next approximation for f_{ij} we have to apply $z(z-QLQ)^{-1}=1+QLQ(z-QLQ)^{-1}$ twice again. Then we should make the same approximation as in the previous case thereby obtaining a new set of coupled equations. In our opinion the second approximation for f_{ij} is better than the first one, because more Q operators are involved and a higher moment is needed.

The simplest approximation one can make for the memory function is to replace it by its value in $i\epsilon$. In fact, one supposes that the memory function decays very fast as a function of time compared to the natural motion of the system.[10,13] In our approach we do the same, but in a later stage, as can be seen from Eqs. (2.7)–(2.9). Then we get an expression for the memory function, where its own value in $i\epsilon$ occurs as a parameter. However, because this approximation for the memory function is frequency dependent, we can use the sum rule Eq. (2.11) to determine this parameter.

As will be seen later on, the sum rules on the memory function also imply sum rules on the relaxation function.

B. Scalar product

It has been found that the scalar product defined by[10]

$$(A,B)=\int_0^\beta \langle e^{\lambda L}A^\dagger B\rangle d\lambda\,, \tag{2.12}$$

where $\langle\ \rangle$ stands for the thermal expectation value, is a very convenient tool to describe dynamical properties. It represents the static susceptibility $\chi_{AB}(0)$, and therefore the function

$$\begin{aligned}\Phi_{AB}(z)&=(A,(z-L)^{-1}B)\\ &=-i\int_0^\infty dt\,e^{izt}\int_0^\beta d\lambda\,\langle e^{\lambda L}A^\dagger(t)B(0)\rangle,\\ &\qquad\qquad \mathrm{Im}z>0\,,\end{aligned} \tag{2.13}$$

corresponds to the Laplace transformed relaxation function. Since the fluctuation dissipation theorem (see Appendix B) provides relations between the spectral density of the symmetrized correlation function and static susceptibilities, it is useful to define another scalar product by

$$(A\,|\,B)=\tfrac{1}{2}\langle\{A^\dagger,B\}\rangle\,, \tag{2.14}$$

where $\{\ ,\ \}$ denotes the anticommutator.

Although this scalar product is less interesting from the physical point of view,[10] we will show that it can be used to calculate certain static quantities.

Therefore we define, in complete analogy with the previous case, the Laplace transformed symmetrized correlation function by

$$\begin{aligned}\psi_{AB}(z)&=(A\,|\,(z-L)^{-1}B)\\ &=-\frac{i}{2}\int_0^\infty dt\,e^{izt}\langle\{A^\dagger(t),B(0)\}\rangle,\quad \mathrm{Im}z>0\,.\end{aligned} \tag{2.15}$$

From these equations, it should be clear that we can apply the theory, given in Sec. II A, to the relaxation function or the symmetrized correlation function, depending on the choice of the scalar product, while the fluctuation dissipation theorem will give us a relation between the static correlation functions and the static susceptibility.

It is well known that the fluctuation dissipation theorem is equivalent to the statement that the system is in thermal equilibrium.[14] Therefore we can correctly account for the fact that we assume that the system is in equilibrium. Of course the expressions for the static susceptibilities will depend on the approximation for the dynamic quantities, but as outlined in the previous section, we can make the approximation systematically better.

III. MODEL

The Hamiltonian for the one-dimensional Heisenberg system with nearest-neighbor interaction is given by

$$H=-J\sum_i \vec{S}_i\cdot\vec{S}_{i+1}\,. \tag{3.1}$$

If J is positive the system is ferromagnetic and for negative J values it is antiferromagnetic. Defining Fourier transformed operators by

$$S_k^\alpha=\frac{1}{\sqrt{N}}\sum_n e^{ikn}S_n^\alpha\,,\quad \alpha=\pm,z\,, \tag{3.2}$$

we have

$$H=-J\sum_k (S_k^+S_{-k}^-+S_k^zS_{-k}^z)\cos k\,. \tag{3.3}$$

Here and in the following, N denotes the number of spins. For simplicity we have chosen the lattice constant equal to one and we use units where $\hbar=1$, $k_B=1$.

Since a one-dimensional Heisenberg magnet is isotropic at finite temperatures, it is sufficient to take one spin component. As the total spin $\vec{S}_{k=0}$ is a conserved quantity, we have $L^n\vec{S}_{k=0}=0$, $n=1,2,\ldots$ and consequently we have to take[10]

$$\{S_k^z, LS_k^z, L^2S_k^z, \ldots\} \tag{3.4}$$

as the set of relevant variables. However, this set is rather large for obvious reasons, and therefore we will not work with a set larger than

$$\{S_k^z, LS_k^z, L^2S_k^z\}\,. \tag{3.5}$$

Because the energy density is a conserved quantity we should treat it as an additional variable but since it couples to the other variables in the transport coefficients only, we neglect it. We close this section with a remark on the notation of the scalar product. If A_k and B_q denote Fourier transformed operators then $(A_k,B_q)=\delta_{kq}(A_k,B_k)$ because of translational invariance. Therefore we will write $(A,B)_k$ in the following. Of course, this also holds for the scalar product defined by Eq. (2.14).

IV. DYNAMIC EQUATIONS

For clarity, we rewrite the basic equation (2.5) for the relaxation matrix $\tilde{\Phi}(z,q)$ in the well-known form[13]

$$[z\tilde{1} - \tilde{\Omega}_q + \tilde{\Sigma}_q(z)]\tilde{\Phi}(z,q) = \tilde{\chi}_q, \tag{4.1}$$

where $\tilde{1}$ represents the unit matrix, $\tilde{\Omega}_q$ the frequency matrix, $\tilde{\Sigma}_q(z)$ the memory matrix, and $\tilde{\chi}_q$ the static susceptibility matrix.

At first we will take

$$\{S_q^z, LS_q^z\} \tag{4.2}$$

as our set of secular variables. In the following we drop the superscript z. In this case the static susceptibility matrix $\tilde{\chi}_q$ and the projector P on the space spanned by the secular variables are given by

$$\tilde{\chi}_q = \begin{pmatrix} (S,S)_q & 0 \\ 0 & (LS,LS)_q \end{pmatrix}, \tag{4.3}$$

and

$$PB_q = \frac{(S,B)_q}{(S,S)_q} S_q + \frac{(LS,B)_q}{(LS,LS)_q} LS_q . \tag{4.4}$$

Here B denotes an arbitrary operator.

The frequency matrix is given by

$$\begin{gathered}\tilde{\Omega}_q = \begin{pmatrix} 0 & (LS,LS)_q \\ (LS,LS)_q & 0 \end{pmatrix}, \\ \tilde{\chi}_q^{-1} = \begin{pmatrix} 0 & 1 \\ \dfrac{(LS,LS)_q}{(S,S)_q} & 0 \end{pmatrix},\end{gathered} \tag{4.5}$$

and because $QLS_q = (1-P)LS_q = 0$, the memory matrix can be written as

$$\begin{aligned}\tilde{\Sigma}_q(z) &= \begin{pmatrix} 0 & 0 \\ 0 & -(QL^2S, (z-QLQ)^{-1}QL^2S)_q \end{pmatrix} \tilde{\chi}_q^{-1} \\ &= \begin{pmatrix} 0 & 0 \\ 0 & \Sigma_q(z) \end{pmatrix},\end{aligned} \tag{4.6a}$$

where

$$\Sigma_q(z) = -(LS,LS)_q^{-1}(QL^2S, (z-QLQ)^{-1}QL^2S)_q . \tag{4.6b}$$

Now Eq. (4.1) reads

$$\begin{pmatrix} z & -1 \\ -\dfrac{(LS,LS)_q}{(S,S)_q} & z+\Sigma_q(z) \end{pmatrix} \begin{pmatrix} \Phi_{SS}(z,q) & \Phi_{S\,LS}(z,q) \\ \Phi_{LS\,S}(z,q) & \Phi_{LS\,LS}(z,q) \end{pmatrix} = \begin{pmatrix} (S,S)_q & 0 \\ 0 & (LS,LS)_q \end{pmatrix}, \tag{4.7}$$

and the solution for $\Phi_{SS}(z,q)$ yields

$$\Phi_{SS}(z,q) = \frac{(S,S)_q[z+\Sigma_q(z)]}{z^2 + \Sigma_q(z)z - (LS,LS)_q/(S,S)_q} . \tag{4.8}$$

In order to show how the general theory, exposed in Sec. II, works, we will now evaluate the transport coefficient $\Sigma_q(z)$ explicitly. Using the identity $z(z-QLQ)^{-1} = 1 + QLQ(z-QLQ)^{-1}$ we find

$$\begin{aligned} z(QL^2S, (z-QLQ)^{-1}QL^2S)_q \\ = (QL^2S, L^2S)_q + (QLQL^2S, (z-QLQ)^{-1}QL^2S)_q ,\end{aligned} \tag{4.9}$$

where we have used Eq. (2.2). Multiplying the equation with z and using the identity once more, we get

$$\begin{aligned} z^2(QL^2S, (z-QLQ)^{-1}QL^2S)_q \\ = z(QL^2S, L^2S)_q + (QLQLQL^2S, (z-QLQ)^{-1}QL^2S)_q ,\end{aligned} \tag{4.10}$$

where the term $(QLQL^2S, QL^2S)_q$ is omitted right away because it is zero by time reversal invariance. It is easy to verify that we can write

$$QLQLQL^2S_q = -\frac{(L^2S, L^2S)_q}{(LS,LS)_q} QL^2S_q + QL^4S_q , \tag{4.11}$$

and so in this particular case, the general Eq. (2.7) becomes

$$\begin{aligned} \left(z^2 + \frac{(L^2S,L^2S)_q}{(LS,LS)_q}\right)(QL^2S, (z-QLQ)^{-1}QL^2S)_q \\ = z(QL^2S, L^2S)_q + (QL^4S, (z-QLQ)^{-1}QL^2S)_q .\end{aligned} \tag{4.12}$$

Following the general theory we replace the second term of the right-hand side of Eq. (4.12) by a complex number R and we have

$$\begin{aligned} \left(z^2 + \frac{(L^2S,L^2S)_q}{(LS,LS)_q}\right)(QL^2S, (z-QLQ)^{-1}QL^2S)_q \\ = z(QL^2S, L^2S)_q + R .\end{aligned} \tag{4.13a}$$

Taking the limit $z = i\epsilon$, $\epsilon \to 0$ we have

$$\frac{(L^2S,L^2S)_q}{(LS,LS)_q} \lim_{z\to i\epsilon} (QL^2S, (z-QLQ)^{-1}QL^2S)_q = R . \tag{4.13b}$$

From symmetry considerations (see Appendix A) we know that $\lim_{z\to i\epsilon}(QL^2S, (z-QLQ)^{-1}QL^2S)_q$ must be pure imaginary. Then Eq. (2.11) reads

$$(QL^2S, L^2S)_q = -\frac{1}{\pi} \operatorname{Im}(R) \int_{-\infty}^{+\infty} d\omega [\omega^2 + (L^2S, L^2S)_q/(LS,LS)_q]^{-1} , \tag{4.14a}$$

and consequently we have

$$\mathrm{Im}(R)=-(QL^2S,L^2S)_q\frac{(L^2S,L^2S)_q^{1/2}}{(LS,LS)_q^{1/2}},\quad \mathrm{Re}(R)=0\,. \tag{4.14b}$$

Substituting this expression into Eq. (4.13a) we finally obtain

$$\Sigma_q(z)=-\frac{1}{(LS,LS)_q}\frac{(QL^2S,L^2S)_q}{z+i(L^2S,L^2S)_q^{1/2}/(LS,LS)_q^{1/2}}\,. \tag{4.15}$$

Now it is clear that we have extracted the slowest varying contributions out of the memory term. Furthermore Eq. (4.15) shows that, in this approximation, the memory term decays exponentially in time, in agreement with Lovesey and Loveluck.[15] However, we do not need additional approximations for the calculations of the relaxation rates in terms of static quantities. Inserting this expression in the equation for the relaxation function Eq. (4.8), and expanding the relaxation function for large z values, we arrive at[13]

$$\Phi_{SS}(z,q)=\frac{(S,S)_q}{z}+\frac{(LS,LS)_q}{z^3}+\frac{(L^2S,L^2S)_q}{z^5}+\cdots\,. \tag{4.16}$$

This means that our relaxation function now satisfies the sum rule Eq. (B10) for $n=0,1,2$. The first two sum rules are fulfilled by construction, because we have taken S_q and LS_q as our secular variables. As far as the third sum rule is concerned, this one seems to be implied by the sum rule on the memory function. Indeed, going one step further in the determination of the memory function, we find that the next sum rule, for $n=3$, is also fulfilled. The quantity of interest for inelastic neutron scattering is the dynamic form factor[16]

$$S(\omega,q)=\frac{-\omega}{1-e^{-\beta\omega}}\Phi''_{SS}(\omega,q)\,, \tag{4.17}$$

where $\Phi''_{SS}(\omega,q)$ represents the imaginary part of the relaxation function. Within the approximations, given earlier, $\Phi''_{SS}(\omega,q)$ looks like

$$\Phi''_{SS}(\omega,q)=-\frac{[(L^2S,L^2S)_q-(LS,LS)_q^2/(S,S)_q](L^2S,L^2S)_q^{1/2}/(LS,LS)_q^{1/2}}{\omega^2[\omega^2-(L^2S,L^2S)_q/(LS,LS)_q]^2+[(L^2S,L^2S)_q/(LS,LS)_q][\omega^2-(LS,LS)_q/(S,S)_q]^2}\,. \tag{4.18}$$

If this approximation for the relaxation function would turn out to be a bad one, there could be two reasons: our set of secular variables is not convenient, or we would have to evaluate $\Sigma_q(z)$ better.

A. Classical limit

To test the expression for the relaxation function we now turn to the classical Heisenberg chain. For this system the statics were solved exactly, and the relation between the static susceptibility and the static correlation function becomes extremely simple:

$$(S,S)_q=\beta(S\,|\,S)_q=\beta\,\langle S_q^zS_{-q}^z\rangle\,. \tag{4.19}$$

As there exists a lot of literature on the classical Heisenberg chain we here merely state the most important results. The static correlation function is given by[5,6,17]

$$\langle S_q^zS_{-q}^z\rangle=\frac{S^2}{3}\frac{1-v^2}{1+v^2-2v\cos q}\,, \tag{4.20a}$$

with

$$v=\coth(\beta JS^2)-1/\beta JS^2\,. \tag{4.20b}$$

Here S stands for $[S(S+1)]^{1/2}$. The classical inverse correlation length, which is a measure for the extention of the short-range order in the system, is written as a function of temperature in the following way:

$$\kappa=-\ln|v|\,. \tag{4.21}$$

The first classical moments were calculated by several authors, and the results for the static susceptibilities are[5,17]

$$(S,S)_q=\beta\langle S_q^zS_{-q}^z\rangle\,, \tag{4.22a}$$

$$(LS,LS)_q=\tfrac{4}{3}JS^2(1-\cos q)v\,, \tag{4.22b}$$

$$\begin{aligned}(L^2S,L^2S)_q=\tfrac{8}{9}J^3S^4(1-\cos q)&\\ \times[5v-2v^2+3y+vy-v^2y&\\ -3v(1+2v+y)\cos q+2v^2(2+y)\cos^2 q]\,,&\end{aligned} \tag{4.22c}$$

with

$$y=1-3v/\beta JS^2\,. \tag{4.22d}$$

In the low-temperature limit, β → ∞, the classical results reduce to

$$\kappa\rightarrow 1/\beta\,|J|\,S^2\,, \tag{4.23a}$$

$$(S,S)_q\rightarrow 1/3\,|J|\,(1\mp\cos q)\,, \tag{4.23b}$$

$$(LS,LS)_q\rightarrow\tfrac{4}{3}\,|J|\,S^2(1-\cos q)\,, \tag{4.23c}$$

$$(L^2S,L^2S)_q\rightarrow\tfrac{16}{3}\,|J|^3S^4(1-\cos q)^2(1\mp\cos q)\,, \tag{4.23d}$$

where the minus sign refers to the ferromagnet.

The dynamic form factor for neutron scattering Eq. (4.17), in the classical limit, reduces to

$$S(\omega, q)=(-1/\beta)\Phi''_{SS}(\omega, q)\,. \tag{4.24}$$

From Eq. (4.18), and the fact that

$$\lim_{\beta\to\infty}\frac{(S,S)_q(L^2S,L^2S)_q}{(LS,LS)_q^2}=1\,, \tag{4.25}$$

we conclude that in this limit, the spectrum consists of two very sharp peaks, symmetric with respect to the origin, and with their positions determined by

$$\Omega(q)=2JS(1-\cos q) \tag{4.26a}$$

in the case of a ferromagnet ($J>0$), and

$$\Omega(q)=2S\,|J\sin q\,| \tag{4.26b}$$

in the case of an antiferromagnet ($J<0$). With increasing temperature, the widths of the peaks increase and their heights decrease.

We will now summarize the main results, as well as the comparison with experiment and other theories. For the ferromagnet, for small wave vectors, we find that well-defined spin waves exist if $q\gg\kappa^{1/2}$, where κ now is given by Eq. (4.23a). A central peak is seen when $q\ll\kappa^{1/2}$. For larger wave vectors and increasing temperature, the spectrum changes from a two-peak structure to a three-peak structure. Of course the heights of the peaks decrease with increasing temperature, as well as the areas under the curve, since they are given by $(1/\beta)(S,S)=\langle S_q^zS_{-q}^z\rangle$. For the antiferromagnet, experimental data are available for TMMC. Because TMMC has spin $\frac{5}{2}$, we expect the classical model to give a reasonable description. The region of greatest experimental interest for the antiferromagnet is not the small-q region, but the region for which q^* is small, where q^* is defined by

$$q^*=\pi-q\,. \tag{4.27}$$

This can easily be understood as follows: because in the antiferromagnet two neighboring spins want to stand opposite one to the other, the probability for observing a spin wave with a certain small-q vector (long wavelength) is much smaller than for observing a spin wave with the corresponding small-q^* vector, although their excitation energies are equal as can be seen from the dispersion relation Eq. (4.26b). We then find for the condition for the existence of spin waves in the small-q^* region: $q^*\gg\kappa^{1/2}$. Our results are in good agreement with theories of Lovesey and Meserve,[4] who use a termination function in a continued fraction method, and of Tomita and Mashiyama,[5] who use a Gaussian assumption. The latest experiments on TMMC,[8] which were done for very small q^* values, however, revealed that the existence of spin waves is not connected with $\kappa^{1/2}$ but with κ. From the Fig. 1 it is obvious that our results cannot predict spin waves for very small q^* values, at the temperatures they are measured.

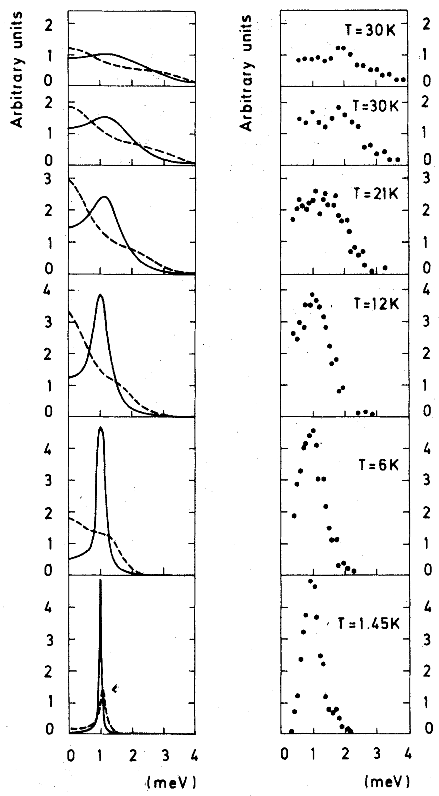

FIG. 1. Theoretical and experimental line shapes for $q^*=0.05\pi$. The dashed lines are the results for the set of variables $\{S_qLS_q\}$, the full lines for $\{S_q, LS_q, A_q\}$. We took $J=-13$ K, $S=\frac{5}{2}$, and only adjusted the theoretical results so that the height of the peak at 12 K is equal to that of the measured one. To make a detailed comparison, one should convolute the theoretical spectral function with the experimental resolution function. The experimental resolution is in this case 0.6 meV, which is equal to the width of the measured peak at $T=1.45$ K, and therefore the spin-wave linewidth is certainly smaller than the experimental one.

As already mentioned, one thing we can do is to improve the determination of the memory function. Then one finds

$$(QL^2S,(z-QLQ)^{-1}QL^2S)_q=\frac{z^3(QL^2S,L^2S)_q+z\,(QL^4S,L^2S)_q-i\lambda_q}{z^4+[(L^2S,L^2S)_q/(LS,LS)_q]z^2+(L^3S,L^3S)_q/(LS,LS)_q}\,, \tag{4.28a}$$

$$\lambda_q=(QL^2S,L^2S)_q\frac{(L^3S,L^3S)_q^{1/2}}{(LS,LS)_q^{1/2}}\left(\frac{(L^2S,L^2S)_q}{(LS,LS)_q}+2\frac{(L^3S,L^3S)_q^{1/2}}{(LS,LS)_q^{1/2}}\right)^{1/2}, \tag{4.28b}$$

where the expression for $(L^3S, L^3S)_q$ is taken from Ref. 5. We found that, doing this, the results were not improved drastically. Therefore we concluded that our set of secular variables was not satisfying.

The next secular variable we can take is L^2S_q. For convenience, however, we will take

$$A_q = L^2S_q - \frac{(LS, LS)_q}{(S,S)_q} S_q , \qquad (4.29)$$

because this choice makes the static susceptibility diagonal. Our set of secular variables is thus $\{S_q, LS_q, A_q\}$. The susceptibility matrix and the projector P are readily found to be

$$\tilde{\chi}_q = \begin{pmatrix} (S,S)_q & 0 & 0 \\ 0 & (LS, LS)_q & 0 \\ 0 & 0 & (A,A)_q \end{pmatrix}$$

$$(A,A)_q = (L^2S, L^2S)_q - (LS, LS)_q^2/(S,S)_q , \qquad (4.30)$$

and

$$PB_q = \frac{(S,B)_q}{(S,S)_q} S_q + \frac{(LS,B)_q}{(LS,LS)_q} LS_q + \frac{(A,B)_q}{(A,A)_q} A_q . \qquad (4.31)$$

Again B denotes an arbitrary operator.

The frequency matrix is also easy to calculate

$$\tilde{\Omega}_q = \begin{bmatrix} 0 & 1 & 0 \\ \dfrac{(LS, LS)_q}{(S,S)_q} & 0 & 1 \\ 0 & \dfrac{(A,A)_q}{(LS, LS)_q} & 0 \end{bmatrix} , \qquad (4.32)$$

and the memory matrix contains one nonzero element

$$\tilde{\Sigma}_q(z) = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \Sigma_q(z) \end{pmatrix} , \qquad (4.33a)$$

with

$$\Sigma_q(z) = -\frac{1}{(A,A)_q} (QLA, (z - QLQ)^{-1} QLA)_q . \qquad (4.33b)$$

The solution of the matrix equation Eq. (4.1) gives for the relaxation function

$$\Phi_{SS}(z,q) = \frac{(S,S)_q[z^2 + \Sigma_q(z)z - (A,A)_q/(LS,LS)_q]}{z[z^2 - (L^2S, L^2S)_q/(LS,LS)_q] + \Sigma_q(z)[z^2 - (LS,LS)_q/(S,S)_q]} . \qquad (4.34)$$

For the memory function we find

$$\Sigma_q(z) = -\frac{1}{(A,A)_q} \frac{(QLA, LA)_q}{z + i(LA, LA)_q^{1/2}/(A,A)_q^{1/2}} , \qquad (4.35a)$$

$$(LA, LA)_q = (L^3S, L^3S)_q - 2\frac{(LS,LS)_q(L^2S, L^2S)_q}{(S,S)_q} + \frac{(LS,LS)_q^3}{(S,S)_q^2} , \qquad (4.35b)$$

$$(QLA, LA)_q = (L^3S, L^3S)_q - \frac{(L^2S, L^2S)_q^2}{(LS,LS)_q} . \qquad (4.35c)$$

We now find that the criterion for the existence of spin waves for small-q values in the ferromagnet and in the antiferromagnet is $q \gg \kappa$. More precisely, the line shape goes over from a two-peak structure to a one-peak structure at $\kappa \simeq 2.5q$ for the ferromagnet and at $\kappa \simeq q$ for the antiferromagnet. If we make a comparison with the experiment[8] for the wave vector $q^* = 0.05\pi$ (Fig. 1), we conclude that we have a much better agreement than with our previous choice of variables. However, especially at the lowest temperature, our linewidth is smaller than the experimental one. We came to the same conclusion for the values $q^* = 0.03\pi$ and $q^* = 0.015\pi$.[8] Generally we have an improvement with respect to our previous results, because they predict a central peak, except at the lowest temperature for $q^* = 0.05\pi$, at the measured temperatures and wave vectors. We also plot the results for the value $q^* = 0.25\pi$ in Fig. 2.

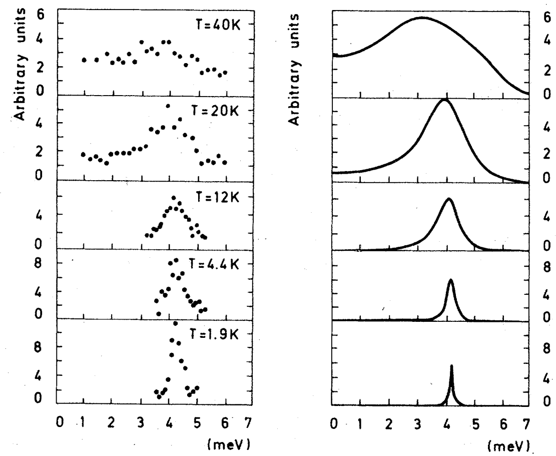

FIG. 2. Theoretical results for $q^* = 0.25\pi$ for the set of variables $\{S_q, LS_q, A_q\}$, compared with experiment. As for Fig. 1 we only adjusted the heights.

We may conclude that in changing the set of secular variables from $\{S_q, LS_q\}$ to $\{S_q, LS_q, A_q\}$, we improve the results considerably. Yet there remains a discrepancy at small-q^* values because our linewidth is too small. The reason is probably that at these small-q^* values and low temperatures, a better determination of the memory function is needed. Of course this is equivalent with knowing higher moments, and therefore at present we are not able to do this. Another important cause of error is that the measured system (TMMC) is of course a quantum-mechanical one, and especially at these low temperatures, we expect the quantum-mechanical effects to become important. The difference between the probability for absorption and emission is represented by the Bose factor in Eq. (4.17). As we already mentioned, in the classical case the Bose factor disappears [Eq. (4.24)], and the spectrum becomes symmetric with respect to the origin.

Another quantity of interest, which we can calculate is the diffusion coefficient. In the high-temperature limit $\beta \to 0$, and for small wave vectors q, the equation for the relaxation function, in both cases, reduces to

$$(z + iDq^2)\Phi_{SS}(z,q) = (S, S)_q \,. \tag{4.36}$$

For our first choice of secular variables, we find

$$D = 0.58J[S(S+1)]^{1/2} \tag{4.37}$$

in good agreement with mode-mode coupling calculations.[9,11] For the secular variables $\{S_q, LS_q, A_q\}$, we have

$$D = 1.08J[S(S+1)]^{1/2} \,. \tag{4.38}$$

This result is in much better agreement with the computer result[12] $D = 1.33J[S(S+1)]^{1/2}$ than the previous one.

B. Quantum system

Let us now turn to the quantum mechanical system. Here the situation is much more complicated because only for infinite and zero temperature some exact results on the static quantities are known.[18] We will use the fluctuation dissipation theorem to calculate these quantities.

From our discussion in Sec. II B it is clear that if we take

$$\{S_q, LS_q\}, \tag{4.39}$$

as the set of relevant variables the symmetrized correlation function corresponding to the relaxation function given by Eq. (4.8) reads

$$\Psi_{SS}(z,q) = \frac{(S|S)_q[z + \hat{\Sigma}_q(z)]}{z^2 + \hat{\Sigma}_q(z)z - (LS|LS)_q/(S|S)_q}\,, \tag{4.40a}$$

with

$$\hat{\Sigma}_q(z) = -\frac{1}{(LS|LS)_q}\,\frac{(QL^2S|L^2S)_q}{z + i(L^2S|L^2S)_q^{1/2}/(LS|LS)_q^{1/2}}\,. \tag{4.40b}$$

Writing the quantities $(LS, LS)_q$, $(LS|LS)_q$, $(L^2S, L^2S)_q$, and $(L^2S|L^2S)_q$ in terms of correlation functions we obtain

$$(S|S)_q = \langle S^z_q S^z_{-q}\rangle\,, \tag{4.41a}$$

$$(LS, LS)_q = \frac{1}{N}\sum_k \,[A(k,q) + A(k,-q)]\langle S^z_k S^z_{-k}\rangle\,, \tag{4.41b}$$

$$(LS|LS)_q = -\frac{1}{4N}\sum_{k,p} A(k,-q)A(p,q)\langle S^+_k S^-_{-k-q} S^+_p S^-_{-p+q}\rangle\,, \tag{4.41c}$$

$$\begin{aligned}(L^2S, L^2S)_q = {}& \frac{1}{4N^2}\sum_{l,k,p}[A(l,-q) + A(l-p+k,q)][A(p,k-q)A(k,q) + A(k,p+q)A(p,-q)]\langle S^+_l S^+_k S^-_{-l+p-k} S^-_{-p}\rangle \\ &+ \frac{1}{2N^2}\sum_{l,k,p}\{A(p,q)[A(l,k+l)A(k+l,q) + A(k,q-l)A(l,q)] + A(l,-q) \\ &\times[A(p,k+l)A(k+l,-q) - A(k+l+q,p+q)A(p,-q)]\}\langle S^+_l S^z_k S^z_{-l+p-k} S^-_{-p}\rangle\,,\end{aligned} \tag{4.41d}$$

$$\begin{aligned}(L^2S|L^2S)_q = {}& \frac{1}{4N^2}\sum_{\substack{k,p\\ l,m}}[A(p,k+q)A(k,-q) + A(k,p-q)A(p,q)] \\ &\times[A(l,m-q)A(m,q) + A(m,l+q)A(l,-q)]\langle S^+_k S^z_{p-k-q} S^-_{-p} S^+_m S^z_{l-m+q} S^-_{-l}\rangle\,,\end{aligned} \tag{4.41e}$$

where $A(k,q)=J(k)-J(k-q)$. In order to obtain Eqs. (4.41) we explicitly used the symmetry of the system.

Furthermore these quantities are related to each other by the fluctuation dissipation theorem [see Eq. (B9)] and we have

$$(S,S)_q=-\frac{1}{\pi}\int_{-\infty}^{+\infty} d\omega\, \frac{2}{\omega}\, \tanh\frac{\beta\omega}{2}\, \Psi''_{SS}(\omega,q)\,, \tag{4.42a}$$

$$(LS,LS)_q=-\frac{1}{\pi}\int_{-\infty}^{+\infty} d\omega\, 2\omega \tanh\frac{\beta\omega}{2}\, \Psi''_{SS}(\omega,q)\,, \tag{4.42b}$$

$$(L^2S,L^2S)_q=-\frac{1}{\pi}\int_{-\infty}^{+\infty} d\omega\, 2\omega^3 \tanh\frac{\beta\omega}{2}\, \Psi''_{SS}(\omega,q)\,. \tag{4.42c}$$

Notice that if we had started here with the corresponding relaxation function, we do not have an equation similar to Eq. (4.42c) because the integral would diverge. However, Eqs. (4.42) are not sufficient to calculate all quantities because the set is not closed. If we would take instead of LS_k all possible combinations of two spin operators as relevant operators we still have to give the six-spin correlation functions in terms of two- and four-spin correlation functions. Therefore we want to express $(LS|LS)_q$ and $(L^2S|L^2S)_q$ in two-spin correlation functions and the simplest thing we can do is to decouple the correlation functions which determine these quantities. Then we can calculate the correlation function self-consistently from Eq. (4.42b) while Eq. (4.42a) and Eq. (4.42c) give the corresponding values for $(S,S)_q$ and $(L^2S,L^2S)_q$, respectively. Instead of solving these equations simultaneously for a number of q values we first used the classical expression for the correlation function [see Eq. (4.20)] and we determined the parameter v. The integrals of Eq. (4.42) were calculated up to ten decimals by using the residue theorem. In Fig. 3 we compare the obtained value for v with the classical one and we conclude that in the high- and medium-temperature region the difference is very small. However as the temperature decreases we see that our value lies below the classical value given by Eq. (4.20b). Then the corresponding inverse correlation length is greater than the classical one. This means that if the classical relaxation function shows a central peak, this quantum-mechanical relaxation function cannot show spin wave peaks. In order to test the analytic form for the quantum-mechanical correlation function we calculated this quantity directly by solving the equations (4.42) and the equation

$$\frac{1}{N}\sum_k \langle S^z_k S^z_{-k}\rangle=\frac{S(S+1)}{3} \tag{4.43}$$

simultaneously for a hundred different q values. Except for the difference in computer time we did not notice any change. From these results, we conclude we cannot decouple the correlation functions and we feel that it is not necessary to take the larger set of variables if one is not able to make better approximations for higher-order spin-correlation function. We could also introduce bosonlike operators which have been used for the quantum-mechanical one-dimensional Heisenberg system in the low-temperature region by Mikeska.[19] At the moment we are working at this problem.

It should also be mentioned that for a three-dimensional system in the high-temperature region, one can approximately calculate higher-order correlation functions by diagram techniques[20] and therefore our scheme can give better results. However, in this case, the integrals over q space become too complicated.

V. CONCLUSIONS

The main aim of the present work was to show how one can calculate frequency-dependent transport coefficients in a very simple and systematic way. We have applied this method to the classical one-dimensional Heisenberg magnet and we compared the theoretical results with the experimental results for TMMC. Taking $\{S^z_q, LS^z_q\}$ as the set of relevant variables our final relaxation function shows the same behavior as those given by various authors. New experiments revealed that the criterion for observing spin-waves is given by $q^* \gg \kappa$ instead of $q^* \gg \kappa^{1/2}$. Following the general scheme we calculated the transport coefficient in a better approximation but since the obtained line shapes did not differ enough from the previous ones we concluded that our set of variables had to be extended. Taking $L^2S^z_q$ as the additional variable we repeated the calculations for the transport coeffi-

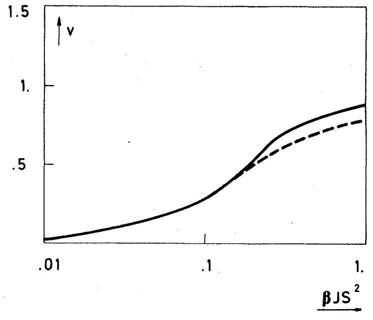

FIG. 3. Comparison between classical value (solid line) and computed value for the parameter v and as a function of inverse temperature.

cients and the relaxation function and, except for a discrepancy in the linewidth, the obtained line shapes are in much better agreement with the experiments than the previous ones.

Observing that the general theory can be used to evaluate the symmetrized correlation function if one introduces another scalar product, we used the fluctuation dissipation theorem to calculate the static quantities for the quantum-mechanical system. In order to obtain a closed set of equations we had to decouple some static correlation functions. From the comparison of our classical results and the less satisfying results obtained for the quantum case we concluded that the decoupling procedure is a fatal approximation. A possible solution for the problem is that one has to introduce boson operators and at present we are working at this problem.

ACKNOWLEDGMENTS

We would like to thank K. H. Michel for suggesting the present investigation. We are grateful to K. H. Michel, Ad Lagendijk, and J. Naudts for useful discussions and to F. Wegner for valuable comments on the manuscript. Finally we thank G. Shirane for sending us experimental results prior to publication.

APPENDIX A

In this Appendix we summarize some properties of the scalar products. In the following A, B, and C are elements of F. By definition, a positive definite scalar product $(A;B)$ must fulfill the following relations:

$$(A;B)^* = (B;A)\,, \tag{A1}$$

$$(A;\lambda B) = \lambda(A;B), \quad \lambda\text{: complex number.} \tag{A2}$$

$$(A;B+C) = (A;B) + (A;C)\,, \tag{A3}$$

$$(A;A) = 0 \Rightarrow A = 0\,. \tag{A4}$$

One easily verifies that the scalar products defined by Eq. (2.12) and Eq. (2.14) satisfy the required conditions. Furthermore they have the remarkable property

$$(A\,|\,B) = (B^\dagger\,|\,A^\dagger); \quad (A,B) = (B^\dagger, A^\dagger)\,. \tag{A5}$$

Remembering the cyclic permutation property in a trace we immediately have

$$(A\,|\,LB) = (LA\,|\,B); \quad (A, LB) = (LA, B) \tag{A6}$$

thereby showing that L is indeed Hermitian.

In general the integral $\int_0^\beta \langle e^{\lambda L} A^\dagger B\rangle\, d\lambda$ cannot be calculated exactly. However, if $B = LC$ we obtain

$$\begin{aligned}(A, LC) &= \int_0^\beta \langle e^{\lambda H} A^\dagger e^{-\lambda H} LC\rangle\, d\lambda \\ &= \int_0^\beta \langle A^\dagger e^{-\lambda H} LC e^{\lambda H}\rangle\, d\lambda \\ &= \int_0^\beta \langle A^\dagger e^{-\lambda H}[H, C] e^{+\lambda H}\rangle\, d\lambda \\ &= -\int_0^\beta d\lambda \frac{d}{d\lambda}\langle A^\dagger e^{-\lambda H} C e^{\lambda H}\rangle \\ &= -\langle A^\dagger e^{-\beta H} C e^{\beta H}\rangle + \langle A^\dagger C\rangle = \langle [A^\dagger, C]\rangle\,.\end{aligned} \tag{A7}$$

After introducing Fourier transformed operators we obtain in a straightforward way

$$(A\,|\,B)_k = (B^\dagger\,|\,A^\dagger)_{-k}; \; (A,B)_k = (B^\dagger, A^\dagger)_{-k}\,. \tag{A8}$$

If the system is invariant under space inversion I we have

$$(A\,|\,B)_k = \epsilon_A^I \epsilon_B^I (A\,|\,B)_{-k}\,, \quad (A,B)_k = \epsilon_A^I \epsilon_B^I (A,B)_{-k}\,, \tag{A9}$$

if $IA(\vec{r})I^\dagger = \epsilon_A^I A(-\vec{r})$, ϵ_A^I denoting the parity of the operator A under space inversion, and $IB(\vec{r})I^\dagger = \epsilon_B^I B(\vec{r})$. If the system is invariant under time inversion we have[21]

$$(A\,|\,B)_k = (\Theta^\dagger B\Theta\,|\,\Theta^\dagger A\Theta)_{-k}; \; (A,B)_k = (\Theta^\dagger B\Theta, \Theta^\dagger A\Theta)_{-k}\,, \tag{A10}$$

where Θ denotes the time inversion operator.

From these equations we can easily deduce similar relations for ψ:

$$\psi_{AB}(z,k) = -\psi_{B^\dagger A^\dagger}(-z,-k) = -\psi^*_{A^\dagger B^\dagger}(-z^*,-k) \tag{A11}$$

and

$$\psi_{AB}(z,k) = \epsilon_A^I \epsilon_B^I \psi_{AB}(z,-k) \tag{A12}$$

if $IA(\vec{r})I^\dagger = \epsilon_A^I A(-\vec{r})$, $IB(\vec{r})I^\dagger = \epsilon_B^I B(-\vec{r})$. The equation obtained by time inversion depends too much on the type of the operators A and B and therefore a general expression is not useful. It is also trivial that Eq. (A11) and Eq. (A12) hold for the relaxation function $\Phi_{AB}(z,k)$.

APPENDIX B

If $\varphi(z)$ is a holomorphic function of z in a domain D of the complex plane and D contains all the eigenvalues of a bounded operator T acting on a Hilbert space E, one defines the operator $\varphi(T)$ by the Dunford-Taylor integral[22]

$$\varphi(T) = \frac{1}{2\pi i}\int_\Gamma \varphi(z)(z-T)^{-1}\, dz\,. \tag{B1}$$

Here $\Gamma \subset D$ may consist of several simple closed curves Γ_k with positive direction, such that the union of the interiors of these curves contains all the eigenvalues of T. If T is Hermitian, the function $(z-T)^{-1}$ has poles for $\mathrm{Im}z = 0$ only. Taking a contour as shown in Fig. 4, with ϵ small and positive, and assuming that $\varphi(z)=\varphi(-z)$ fulfills the required conditions, we easily find

$$\varphi(T)=\frac{-1}{2\pi i}\lim_{\epsilon\to 0}\int_{-\infty}^{+\infty} d\omega\,\varphi(\omega)[(\omega+i\epsilon-T)^{-1} - (\omega-i\epsilon-T)^{-1}]\,. \tag{B2}$$

In a number of cases one can define a similar operator valued function for unbounded operators. Then we have

$$(A\,|\,\varphi(L)B)_k=-\frac{1}{2\pi i}\lim_{\epsilon\to 0}\int_{-\infty}^{+\infty} d\omega\,\varphi(\omega)[\psi_{AB}(\omega+i\epsilon,k) - \psi_{AB}(\omega-i\epsilon,k)]\,. \tag{B3}$$

Using Eq. (A11) we obtain

$$(A\,|\,\varphi(L)B)_k=\frac{-1}{2\pi i}\lim_{\epsilon\to 0}\int_{-\infty}^{+\infty} d\omega\,\varphi(\omega)[\psi_{AB}(\omega+i\epsilon,k) - \psi_{BA}^{*}(\omega+i\epsilon,k)]\,. \tag{B4}$$

For $A=B$ this equation reduces to

$$(A\,|\,\varphi(L)A)_k=-\frac{1}{\pi}\int_{-\infty}^{+\infty} d\omega\,\varphi(\omega)\psi_{AA}''(\omega,k)\,, \tag{B5}$$

where $\psi_{AB}''(\omega,k)$ denotes the imaginary part of $\lim_{\epsilon\to 0}\psi_{AB}(z,k)$. If we take $\varphi(\omega)=\omega^{2n}, n=0,1,\ldots$ we have

$$(L^nA\,|\,L^nA)_k=-\frac{1}{\pi}\int_{-\infty}^{+\infty} d\omega\,\omega^{2n}\psi_{AA}''(\omega,k)\,. \tag{B6}$$

This is the well-known expression for the moments. For $\varphi(\omega)=(1/\omega)\tanh\frac{1}{2}\beta\omega$, Eq. (B4) yields

$$\left(A\,|\,L^{-1}\tanh\frac{\beta L}{2}B\right)_k =-\frac{1}{2\pi i}\int_{-\infty}^{+\infty}\frac{d\omega}{\omega}\tanh\frac{\beta\omega}{2}[\psi_{AB}(\omega+i\epsilon,k) -\psi_{BA}^{*}(\omega+i\epsilon,k)]\,. \tag{B7}$$

On the other hand we have

$$\left(A\,|\,L^{-1}\tanh\frac{\beta L}{2}B\right)_k=\tfrac{1}{2}\langle\{A^{\dagger},L^{-1}(e^{\beta L}-1)(e^{\beta L}+1)^{-1}B\}\rangle_k \tag{B8a}$$

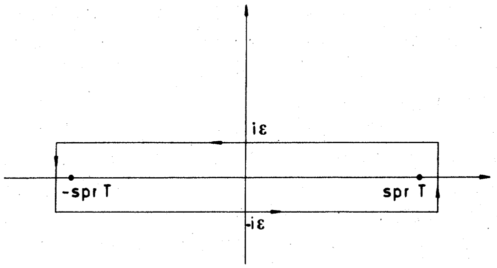

FIG. 4. Integration contour in the complex z plane. Spr$T$ denotes the spectral radius of the operator T.

$$=\frac{1}{2}\int_0^{\beta} d\lambda\,\langle\{A^{\dagger},e^{\lambda L}(e^{\beta L}+1)^{-1}B\}\rangle_k\,. \tag{B8b}$$

Using $\langle A^{\dagger}B\rangle_k=\langle Be^{-\beta L}A^{\dagger}\rangle_k=\langle e^{\beta L}BA^{\dagger}\rangle_k$ repeatedly we find

$$2\left(A\,|\,L^{-1}\tanh\frac{\beta L}{2}B\right)_k=\int_0^{\beta}\langle e^{\lambda L}A^{\dagger}B\rangle_k d\lambda=(A,B)_k\,. \tag{B8c}$$

Consequently we reobtain a fluctuation dissipation theorem[14]

$$(A,B)_k=-\frac{1}{\pi i}\int_{-\infty}^{+\infty}\frac{d\omega}{\omega}\tanh\frac{\beta\omega}{2}[\psi_{AB}(\omega+i\epsilon,k) -\psi_{BA}^{*}(\omega+i\epsilon,k)]\,. \tag{B9}$$

By interchanging the scalar product in Eqs. (B3)–(B9), the equivalents of Eqs. (B6) and (B9) are found to be

$$(L^nA,L^nA)_k=-\frac{1}{\pi}\int_{-\infty}^{+\infty} d\omega\,\omega^{2n}\Phi_{AA}''(\omega,k) \tag{B10}$$

and

$$(A\,|\,B)_k=-\frac{1}{4\pi i}\int_{-\infty}^{+\infty} d\omega\,\omega\coth\frac{\beta\omega}{2}[\Phi_{AB}(\omega+i\epsilon,k) -\Phi_{BA}^{*}(\omega+i\epsilon,k)]\,. \tag{B11}$$

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*Supported by the project "Neutron Scattering," I.I.K.W., Belgium.

†Supported by SFB Ferroelektrika der Deutschen Forschungsgemeinschaft.

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