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# Physical Representation of Spectral Line Shape Based on Moments

Kazuhisa TOMITA and Hiroyuki MASHIYAMA

*Department of Physics, Faculty of Science, University of Kyoto, Kyoto*

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It is pointed out that, depending on the nature of the method of observation, full details of dynamics are not really needed in order to reproduce the canonical correlation spectra. In this spirit a physical representation of a spectrum is proposed on the basis of a few lower-order moments. It may be called the ***n-th order description***, according to the extent to which a reliable knowledge on moments is at hand. As an example the third-order description is analysed in general, assuming the highest stage residual correlation as Gaussian. It is applied to the neutron inelastic scattering spectra in magnetic materials. In the case of an antiferromagnetic Heisenberg chain the results practically coincides with that of computer simulation, and are in good agreement with observed data in TMMC. Calculation of the eighth moment indicates that there will be no qualitative change by including this additional information.

## § 1. Introduction

Connected with the anomalous behaviour of the fluctuation around the second-order phase transition, the scaling theory is now being extended even to the problem of dynamics; however, the whole of the scaling theory is concerned with the overall "spread" of the spectrum. Relatively few theoretical investigations have been published on the detailed "shape" of the response spectrum, which characterizes the observed data on an actual situation in a unique way.

In order to treat this kind of problem, the usual stochastic type approach has an admitted deficiency, for it applies only to the long-time, well-moderated behaviour of a system, namely to only the central part of the spectrum. It is obvious that the behaviour of a moderate wing is relevant in the discussion of line shape. In other words some reliable informations are needed when one is interested in relatively short-time behaviour of the correlation under consideration. The weakness of the stochastic approach lies in that it often violates conservation laws of moments, which is characteristic of the short-time behaviour.

In our previous publications[1),2)] an alternative approach was proposed which does not violate the sum rule to certain orders. Using the continued fraction representation of the canonical correlation spectra of the magnetic moments, and using theoretical estimates of moments up to the fourth, we discussed the spectral shape of neutron inelastic scattering in $RbMnF_3$[3)~5)] in the paramagnetic neighbourhood of the Néel point. In looking into the behaviour of higher order correlations, an assumption was then[1] introduced that the spectral shape of the residual torque on the magnetic moment is *Gaussian.* The computed results based on this assumption was in essential agreement with the observation in so far as the overall width is concerned; however, there remained a discrepancy in the detailed line shape. Namely, in contrast to the three peaks observed the theory predicted only two, and the origin of this discrepancy was traced back to the assumption of simple Gaussian torque. In trying to improve the theory one "step down" was made from the observed data with respect to the continued fraction representation of the canonical correlation. It was then found that there exists an wiggling in the torque correlation, namely the correlation is characterized by an oscillatory damping rather than a monotonic damping as in the case of Gaussian decay.

It is attempted in this paper, therefore, to extend the formalism in such a way that an adequate description is provided, by including the knowledge of moments up to the sixth. The capacity of the formalism has been demonstrated in the case of one-dimensional antiferromagnetic chain and in other cases. As the described kind of wiggling is generally expected, the formalism developed here seems to be of wider use.

In § 2 the general idea and several examples of stepping down the continued fraction are described. On the basis of this experience a stepping-up process is introduced in §§ 3 and 4, assuming the third-order correlation as Gaussian. The resulting spectral line shape is presented as a function of the relative magnitude of three quantities $\Delta_1^2$, $\Delta_2^2$, $\Delta_3^2$, which are functions of moments up to sixth order. In § 5 the theory is applied to some typical examples including one-dimensional chain. In § 6 the plausibility of the assumption adopted in § 4 is discussed on the basis of knowledge on still higher order moments. Several comments are also made on previous works along similar lines.

## § 2. The *n*-th order measurement

The observed inelastic scattering of neutron in a spin system is directly related to the spin pair correlation function. In particular the diffuse part of the cross section[6] is essentially proportional to the real part of the Laplace transform $X^{\alpha\beta}(k,\omega)$ of the canonical correlation function[7] $\Phi^{\alpha\beta}(k,t)$; i.e.,

$$X^{\alpha\beta}(k,\omega)=\int_0^\infty dt\, e^{-i\omega t}\Phi^{\alpha\beta}(k,t)$$
$$=X'^{\alpha\beta}(k,\omega)-iX''^{\alpha\beta}(k,\omega)\ , \tag{1}$$

where

$$\Phi^{\alpha\beta}(k,t)\equiv\langle S^\alpha(k,0)\,;S^\beta(-k,t)\rangle$$
$$=\int_0^\beta d\lambda\{\langle e^{\lambda\mathcal{H}}S^\alpha(k,0)e^{-\lambda\mathcal{H}}S^\beta(-k,t)\rangle-\langle S^\alpha(k)\rangle\langle S^\beta(-k)\rangle\}\ . \tag{2}$$

The canonical correlation spectrum, in its normalized form $\overline{X}^{\alpha\beta}(k,\omega)$, may be written as

$$\overline{X}(k,\omega)=\frac{X(k,\omega)}{\Phi(k,0)}=\frac{1}{i\omega+\Delta_1{}^2\overline{X}_1(k,\omega)}, \tag{3}$$

where $\overline{X}_1(k,\omega)$ is a normalized canonical correlation spectrum of the residual force of first order, i.e.,

$$\overline{X}_1(k,\omega)=\int_0^\infty dt\, e^{-i\omega t}R_1(k,t)$$

and

$$R_i(k,t)=\Phi_i(k,t)/\Phi_i(k,0)\ .$$

Here $\Phi_1(k,t)$ is the correlation function $\langle\dot{S}_\perp(k,0);\dot{S}_\perp(-k,t)\rangle$, where $\dot{S}_\perp(-k,t)$ is the part of $\dot{S}(-k,t)$ which is orthogonal to $S(-k)$.[8] It is clear that this procedure can be repeated, and one finds

$$\overline{X}_{i-1}(k,\omega)=\frac{1}{i\omega+\Delta_i{}^2\overline{X}_i(k,\omega)}\ . \tag{4}$$

Separating the real and the imaginary parts, one finds a recurrence type relation

$$\overline{X}'_{i-1}=\frac{\Delta_i{}^2\overline{X}_i{}'}{(\omega-\Delta_i{}^2\overline{X}_i{}'')^2+(\Delta_i{}^2\overline{X}_i{}')^2}, \tag{5a}$$

$$\overline{X}''_{i-1}=\frac{\omega-\Delta_i{}^2\overline{X}_i{}''}{(\omega-\Delta_i{}^2\overline{X}_i{}'')^2+(\Delta_i{}^2\overline{X}_i{}')^2}, \tag{5b}$$

or conversely

$$\overline{X}'_{i+1}=\frac{1}{\Delta_{i+1}^2}\,\frac{\overline{X}_i{}'}{(\overline{X}_i{}')^2+(\overline{X}_i{}'')^2}, \tag{6a}$$

$$\overline{X}''_{i+1}=\frac{1}{\Delta_{i+1}^2}\left\{\omega-\frac{\overline{X}_i{}''}{(\overline{X}_i{}')^2+(\overline{X}_i{}'')^2}\right\}. \tag{6b}$$

It is well known that $\Delta_i{}^2$ may be expressed in terms of moments up to the $2i$-th order,[9] i.e.,

$$\begin{aligned}&\Delta_1{}^2=m_2,\quad \Delta_2{}^2=[m_4-(m_2)^2]/m_2\,,\\ &\Delta_3{}^2=[m_6m_2-(m_4)^2]/[m_2(m_4-m_2{}^2)]\,,\\ &\cdots,\end{aligned} \tag{7}$$

where $m_i$ stands for the $i$-th moment with respect to $\mathrm{Re}\,X(k,\omega)$.

In the case of a response against an electromagnetic field, what is directly observed is the complex susceptibility

$$\chi(k,\omega)=\chi'(k,\omega)-i\chi''(k,\omega)\ ,$$

which is the Laplace transform of the response function

$$\chi^{\alpha\beta}(k,\omega)=\int_0^\infty dt e^{-i\omega t}\phi^{\alpha\beta}(k,t) .$$

Here $\phi^{\alpha\beta}(k,t)$ is the response function defined by

$$\phi^{\alpha\beta}(k,t)\equiv-\frac{d\Phi^{\alpha\beta}(k,t)}{dt}=\frac{1}{i\hbar}\langle[S^{\alpha}(k,0),S^{\beta}(-k,t)]\rangle .$$

The last relation, when expressed in the frequency domain, takes the following form:

$$\bar{\chi}'(k,\omega)=1-\omega\bar{X}''(k,\omega) , \tag{8a}$$

$$\bar{\chi}''(k,\omega)=\omega\bar{X}'(k,\omega) , \tag{8b}$$

where a normalized susceptibility

$$\bar{\chi}(k,\omega)\equiv\chi(k,\omega)/\Phi(k,0)=\chi(k,\omega)/\chi(k,0)$$

has been introduced.

Using the complex susceptibility, Cole and Cole[10] introduced a very convenient plot for the discussion of line shape. They plotted $\chi''(\omega)$ againt $\chi'(\omega)$, so that each point on the curve corresponds to a particular value of $\omega$. In this plot the change in the overall scale of the spectrum leads to the change only in the rate at which the representative point moves on the locus as a function of $\omega$, and the shape of the locus itself is a *scale independent* representation of the line *shape*. This is the reason why the Cole-Cole plot is particularly suited for the discussion of line shape.

Combining (8) and (6), the relation between the two levels is written in terms of $\chi'$ and $\chi''$,

$$\bar{\chi}'_{i+1}=1-\frac{\omega^2}{\Delta^2_{i+1}}\left\{1-\frac{1-\bar{\chi}_i'}{(1-\bar{\chi}_i')^2+(\bar{\chi}_i'')^2}\right\} , \tag{9a}$$

$$\bar{\chi}''_{i+1}=\frac{\omega^2}{\Delta^2_{i+1}}\left\{\frac{\bar{\chi}_i''}{(1-\bar{\chi}_i')^2+(\bar{\chi}_i'')^2}\right\} . \tag{9b}$$

Giving a correlation spectrum of a quantity $A$ (say polarization) with certain accuracy, and a reliable estimate of a relevant moment, i.e., the second moment with respect to the given spectrum itself, one can always proceed to step down the continued fraction to see the correlation spectrum of $\dot{A}_{\perp}$ (say the residual torque), more precisely the correlation of the residual part of $\dot{A}$.

Repeating this process, one may step down the ladder to any order, and reach an information corresponding to that order. Suppose for example the spectrum is *known* to be exactly *Gaussian*, then in principle all the moments are known as functions of the second moment. This means that in this case one can step down the ladder to infinite order, which brings us to a proto-Gaussian type simple

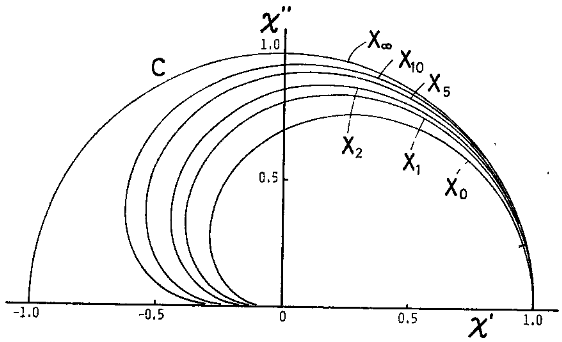



Fig. 1. The Cole-Cole plot of the Gaussian spectrum $X_0(\omega)$ and higher order spectrum $X_i(\omega)$ derived from $X_0(\omega)$.

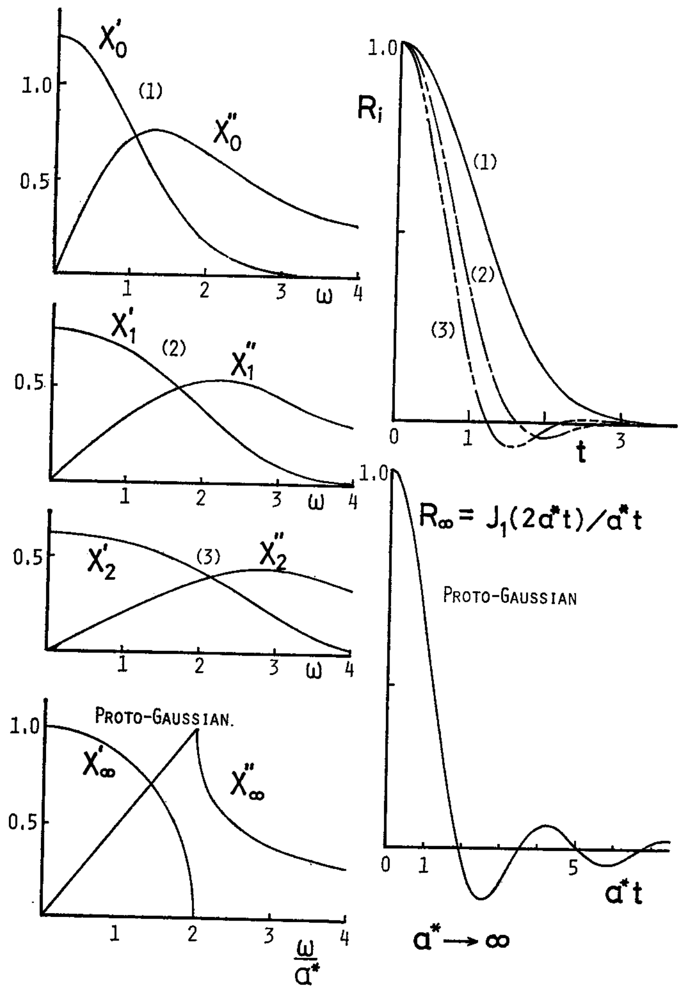
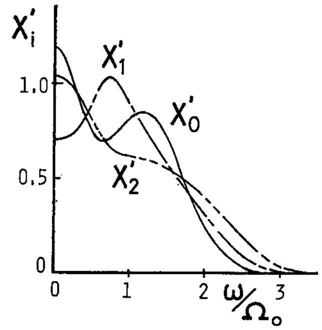



Fig. 2. The canonical correlations $R_i(t)$ and their corresponding spectra $X_i(\omega)$. ($J_1(x)$ is the Bessel function of first order.)

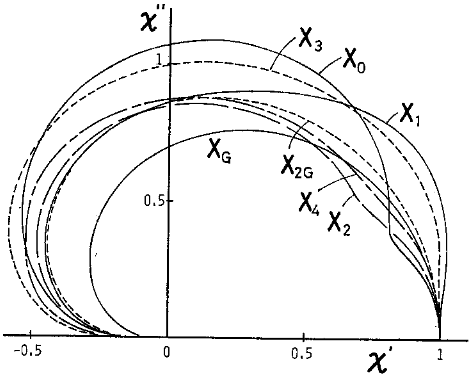
distribution as is shown in Figs. 1 and 2 (cf. the Appendix).

In an actual situation, however, the process cannot be repeated indefinitely because of the difficulties met in the estimation of moments. It is known that one can estimate only the first few moments theoretically, apart from exceptional cases. On an experimental side there is also a difficulty of the noise, which masks the information contained in the wing of the spectrum. As the higher moments are determined almost entirely by the wing, the existence of the noise refuses to provide us with information on the moments due to the system proper beyond a certain order. In practice, therefore, the essential accuracy of the measurement may be thought as limited to the first $n$ moments, which one might call *n-th order measurement.*

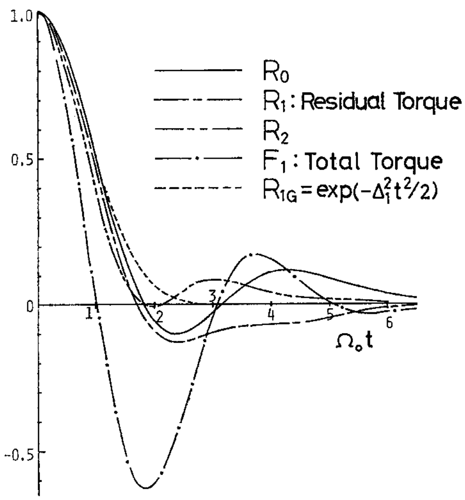
As a typical example of the stepping-down procedure the case of $RbMnF_3$[4] has been taken up. The observed data $X'(k,\omega)$ was first converted into $\Phi(k,t)$, then into $X''(k,\omega)$. The information on $X'(k,\omega)$ and $X''(k,\omega)$ enables us to step down to $X_1'(k,\omega)$ and $X_1''(k,\omega)$ by way of Eq. (6). Repeated use of this formula successively provides us with $X_2(k,\omega)$, $X_3(k,\omega)$ and so forth. At each




(a)




(b)




(c)

Fig. 3. Analytical fit of the experimental data for $RbMnF_3$ (Ref. 4)). (By a superposition of three Gaussian distribution.)

(a) Observed spectrum $X_0'(\omega)$, and its derived spectrum $X_i'(\omega)$. ($\Omega_0^2=(2/3)zS(S+1)(4J^2/\hbar^2)$, $z$: number of nearest neighbours.)

(b) The Cole-Cole plot of $X_i(\omega)$. (Purely Gaussian distribution $X_G(\omega)$ and secondly derived from Gaussian $X_{2G}(\omega)$ are shown for comparison.)

(c) Time correlation $R_i(t)$.

step the canonical correlation functions $\Phi_i(k,t)$ are calculated. The results of these "stepping down" are shown in Fig. 3. In Fig. 3 both the spectrum and the corresponding time correlation function are plotted for $k=(\pi/2, \pi/2, \pi/2)$ at a fixed temperature $T=1.25T_N$. The Cole-Cole plot is helpful in recognizing the reduced line-shape.

It is clear that the total torque correlation

$$F_1(t)=\langle \dot{S}(k,0); \dot{S}(-k,t)\rangle$$

has a large wiggling. Although the wiggling in the residual torque correlation $R_1(t)$ is much less, $R_1(t)$ has still an appreciable negative value range, so that the Gaussian approximation

$$R_{1G}(t)=\exp(-\Delta_1^2t^2/2) ,$$

which was adopted in previous papers has an obvious discrepancy, i.e., about 30% overestimating the area under the correlation function (Fig. 3(c)).

In terms of frequency plot (Fig. 3(a)) the number of peaks in the spectrum is decreased in the process of stepping down, and at the third stage the spectrum becomes one of the type which has single peak at the centre. From the third stage on the higher order correlation spectra have a tendency of convergence to a Gaussian, or perhaps to *pre-Gaussian* line shapes (cf. Fig. 3(b)). The Gaussian decay which turned out too simple for the second-order correlation might be justified as an approximation of the third-order correlation.

It should also be noted that the shape of all even-order correlation $X_{2n}(\omega)$ are close to each other in the relatively low frequency range. The same statement is true among correlation of odd-order $X_{2n+1}(\omega)$; however, there exists a definite difference in shape between $X_{2n}(\omega)$ and $X_{2n+1}(\omega)$. In fact $X_{2n}(\omega)$ is characterized by a marked central peak (cf. Figs. 3(a) and (b)).

## § 3. The *n*-th order description

The concept of $n$-th order measurement leads naturally to the corresponding concept of *n-th order description*. Suppose that the measurement itself is giving reliable information only on the first $2n$ moments of the system proper, in the sense that information on the moments of order higher than $2n$ is blurred by a noise of unknown nature. Then there seems to be little point of worrying about estimation of higher-order moments theoretically. Instead it seems more appropriate to construct an approximate description which may be reliable up to the corresponding order. In other words one may start from a provisional correlation function at the deepest level, and try to "step up" the ladder of continued fraction with reliable estimate of lower-order moments to reach the quantity observed.

In order to step up one may resort to a relation similar to the case of stepping down, i.e., (5a) and (5b), or in terms of complex susceptibility,

$$\chi'_{i-1} = 1 - \frac{\omega^2}{\Delta_i^2} \frac{\omega^2/\Delta_i^2 + \chi_i' - 1}{(\omega^2/\Delta_i^2 + \chi_i' - 1)^2 + (\chi_i'')^2}, \tag{10a}$$

$$\chi''_{i-1} = \frac{\omega^2}{\Delta_i^2} \frac{\chi_i''}{(\omega^2/\Delta_i^2 + \chi_i' - 1)^2 + (\chi_i'')^2}. \tag{10b}$$

Suppose the moments are conserved up to the $2n$-th in order, and the $2(n+1)$-th moment is the lowest moment that is not conserved, then the description may be called "$n$-th order description".

What order of description does one need in a concrete case? Without the foregoing consideration one might imagine that one should proceed to highest possible order. In fact it is not necessary. The answer is obviously dependent on the nature and accuracy of the measurement under consideration. The fact that ordinary measurements are bound to be macroscopic suggests that almost always a finite-order description suffices to cope with the measurement. In this sense the theory of measurement duly entails a stochastic character. The only relevant question is, then, "What order description will be *appropriate* to be compared with a particular type of measurement?"

In the measurement of neutron scattered inelastically by magnetic materials the spectrum exhibits three peaks or two modes. The peak at $\omega=0$ corresponds to a diffusive mode which has practically no restoring force, but the peak at finite frequency indicates the existence of a propagating mode with some kind of restoring force. In the second-order description which was adopted in previous work one of the above two modes appeared at the expense of the other; namely the line shape had either one peak at the centre or two peaks off the centre. The fact that experimentally one can recognize three peaks at the same time means that the second-order description is not really adequate, and one has to proceed a step further. Therefore it is proposed here to adopt the third-order description in order to cope with the coexistence of two modes, or three peaks. Suppose one is content with locating just two modes, and does not care about any more details of the spectrum, then the third-order description will be enough.

## § 4. The spectrum in the third-order description

As in the present situation one is not interested in any detailed structure which is higher than the third in order, it is proposed to assume the third-order residual correlation $R_2(t)$ as a simple Gaussian function

$$R_2(t) = \exp(-\Delta_3^2 t^2/2)$$

or

$$\overline{X}_2'(k, \omega) = \sqrt{\pi/2}\, \exp(-\omega^2/2\Delta_3^2)/\Delta_3,$$

and with this to step up the continued fraction utilizing the knowledge of $\Delta_2$

and $\Delta_1$. This means that the knowledge of moments up to the sixth in order is needed.

Before proceeding to concrete examples it seems appropriate to discuss the general behaviour of third-order description as a function of the small number of parameters involved, provided they are already known. Supposing that one is interested in the shape only, one can scale the frequency by the quantity $\Delta_3$, thus finding the following two essentially independent parameter:

$$a^2 \equiv \Delta_1^2/\Delta_3^2 \quad \text{and} \quad b^2 \equiv \Delta_2^2/\Delta_3^2 .$$

Every possible shape of the spectrum $X'(k, \omega)$ may be uniquely represented by these two parameters. In Fig. 4 a spectral phase diagram is given in $(a^2, b^2)$ plane.

Figure 4 was written by tracing the extremum of the spectral function $X'(\omega)$, namely for a fixed $\omega$ or $\zeta \equiv \omega/\Delta_3$,

$$\frac{\partial}{\partial \omega} X'(\omega; a^2, b^2) = 0$$

was converted into a relation

$$b^2 = G(a^2) .$$

This curve corresponds either to the maximum or the minimum of the spectrum and is shown in Fig. 4 as parametrized by a reduced frequency $\zeta$. The spectral phase boundary corresponds to a situation in which

$$\frac{\partial^2}{\partial \omega^2} X'(\omega; a^2, b^2) = 0$$

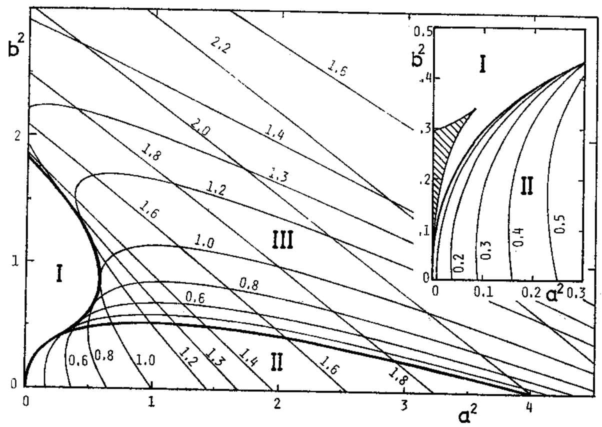



Fig. 4. Spectral phase diagram for third-order description $(R_2(t) = R_G(t))$. Domains I, II, III correspond to single, double and triple peaks, respectively. The loci of extremum are shown with the values of parameter $\zeta = \omega/\Delta_3$.

holds at the same time, namely the maximum turns into the minimum exactly at this point. In this way one can draw the phase boundary in Fig. 4.

The entire region is divided into three domains according to the expected number of peaks; i.e., domains I, II, III correspond to the spectral phase with single, double and triple peaks, respectively. Line shapes and Cole-Cole plots at typical points on the phase diagram are shown in Figs. 5 and 6.

In the Cole-Cole plot it is recognized that line shapes of spectra with three

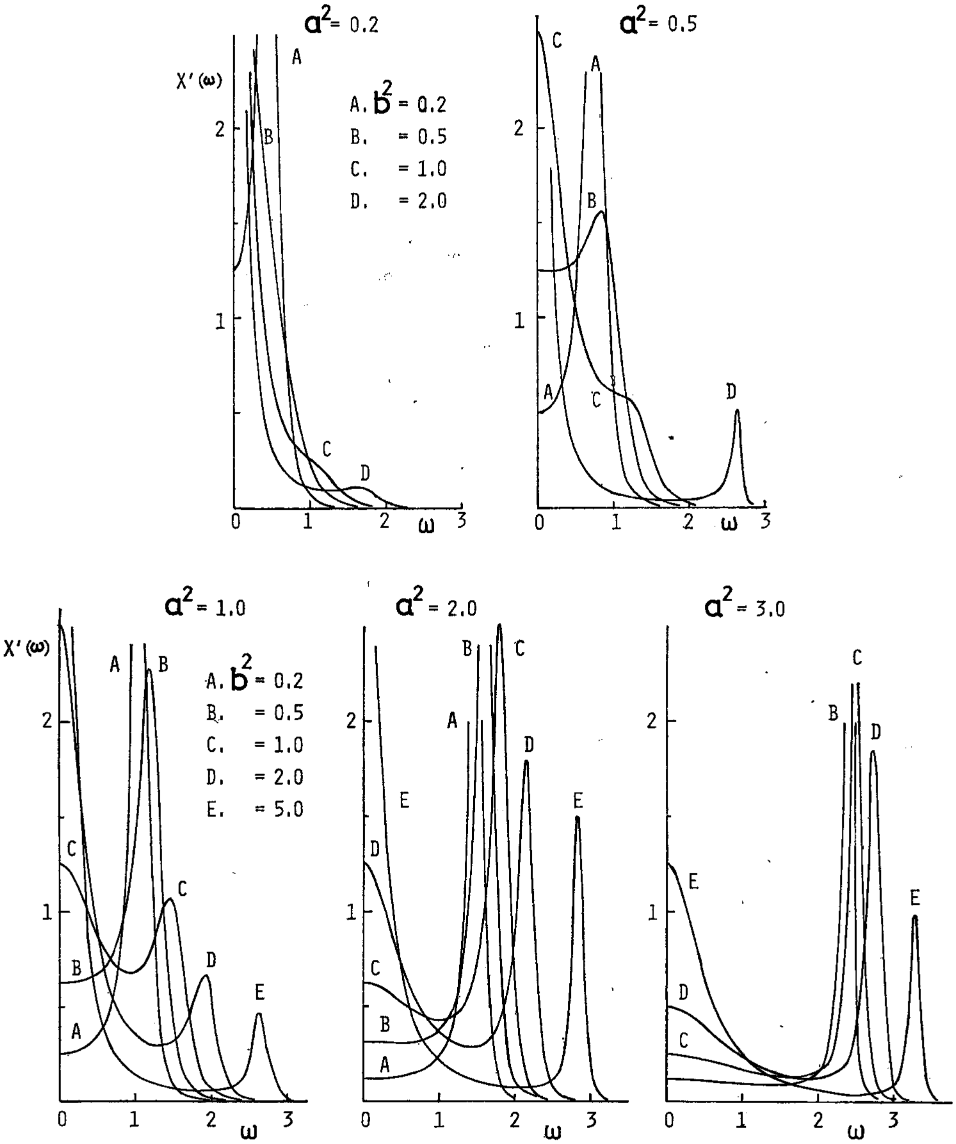



Fig. 5. Line shape at typical points on the phase diagram (Fig. 4).

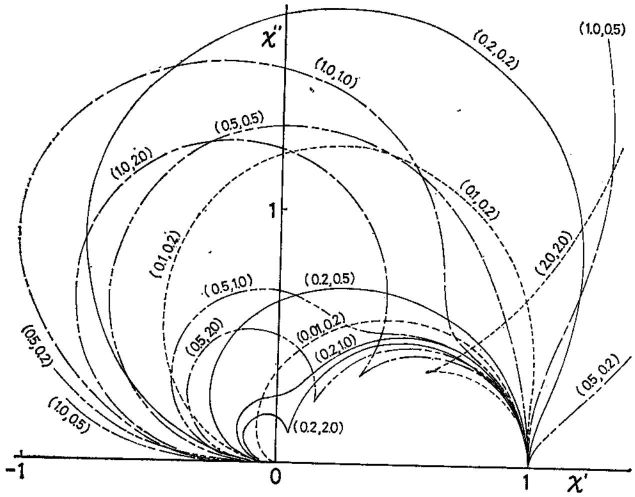



Fig. 6. The Cole-Cole plot corresponding to typical points ($a^2$, $b^2$) on the phase diagram (Fig. 4).

peaks are similar to those with one peak at least in the low frequency range, and are in marked contrast with those with two peaks.

There are three obvious extreme cases in which the value of $\Delta_3$ does not explicitly appear in the shape of the spectrum:

(i) $\overline{X}'(\omega)\sim\pi\delta(\omega)$ for $\Delta_1\ll\Delta_2$, (asymptotic width $\Delta_1^2/\Delta_2$)

(ii) $\overline{X}'(\omega)\sim\frac{\pi}{2}[\delta(\omega-\Delta_1)+\delta(\omega+\Delta_1)]$ for $\Delta_1\gg\Delta_2\sim 0$,

(iii) $\overline{X}'(\omega)\sim\frac{\pi}{\omega_0^2}\left\{\Delta_2^2\delta(\omega)+\frac{\Delta_1^2}{2}[\delta(\omega-\omega_0)+\delta(\omega+\omega_0)]\right\}$ for $\Delta_1, \Delta_2\gg\Delta_3\sim 0$,

where $\omega_0^2=\Delta_1^2+\Delta_2^2$.

It is instructive to analyse the behaviour of poles of the complex function $X(k, \omega)$ in the complex $\omega$ plane. In general there are three poles in this approximation. One is purely imaginary and other two are complex with equal imaginary parts and with real parts which are opposite in sign. The analysis was made and the results are summarized in Fig. 7. The arrows attached to the locus of poles indicate the change induced by an increase in the parameter $a^2$ or $b^2$. The triangular shaded region in Fig. 4 corresponds to the situation in which all the real parts of the poles vanish, and there appear three purely imaginary poles.

It should be noted that this region is definitely smaller than the region I in Fig. 4, because the existence of finite imaginary parts has an effect of merging the contributions from separated poles when they are situated closer to each other compared with the width induced by damping.

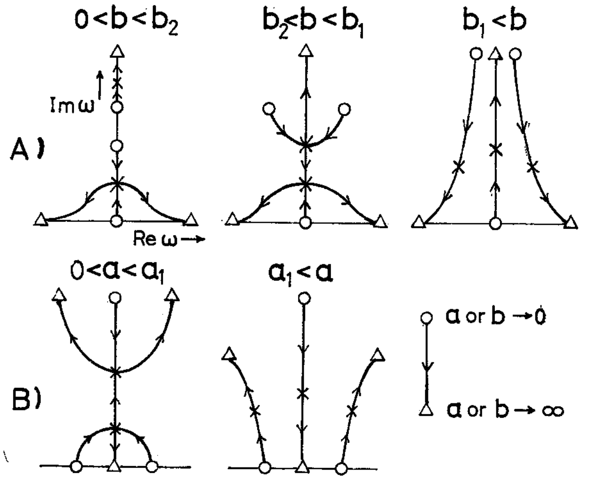



Fig. 7. The position and behaviour of poles for third-order description (schematic). ((A): $b$=constant, (B): $a$=constant; $(a_1^2, b_1^2)$: cusp point, $(0, b_2^2)$: upper bound on $b^2$-axis of the shaded region in Fig. 4.)

There are cases in which the half-value width of the single peak in region I is needed for physical discussion. In Fig. 8 the half-value width $\gamma_{1/2}$ is plotted as a function of $a^2$ and $b^2$. One may recognize that $\gamma_{1/2}$ decreases with decreasing $a^2$ and with increasing $b^2$.

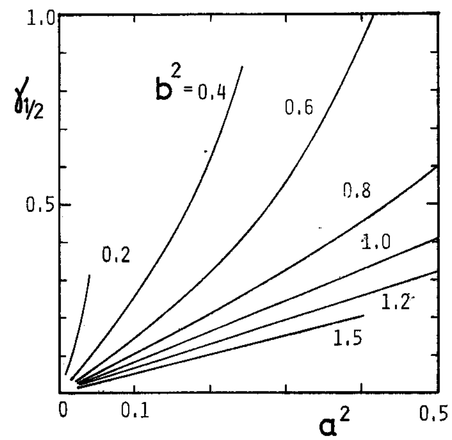



Fig. 8. Half-value width $\gamma_{1/2}$ (third-order description).

## § 5. Applications

In an actual application of third-order description the knowledge of moments up to sixth order is presupposed. It is not, however, an easy task to calculate these moments at general temperature $T$ and for an arbitrary wave number $k$.

The only case in which this was performed is the case of classical linear chain of vector spins, and the present method was successfully applied by our group[2)] to obtain the neutron inelastic scattering in TMMC.[11)] As the details are already published, only the essential results are presented here.

In Fig. 9 contours of constant $k$ and constant $T$ are drawn both for ferromagnets and antiferromagnets. One may then trace the map to find the line shape at an arbitrary temperature and for an arbitrary wave number on this map.

It should be noted that in the high temperature limit the line shape of a ferromagnet for an arbitrary $k$ is identical with that of an antiferromagnet for $k^*=1-k$. In the low temperature limit, however, there exists a marked contrast in shape between ferro- and anti-ferromagnets except for $k=k^*=1/2$.

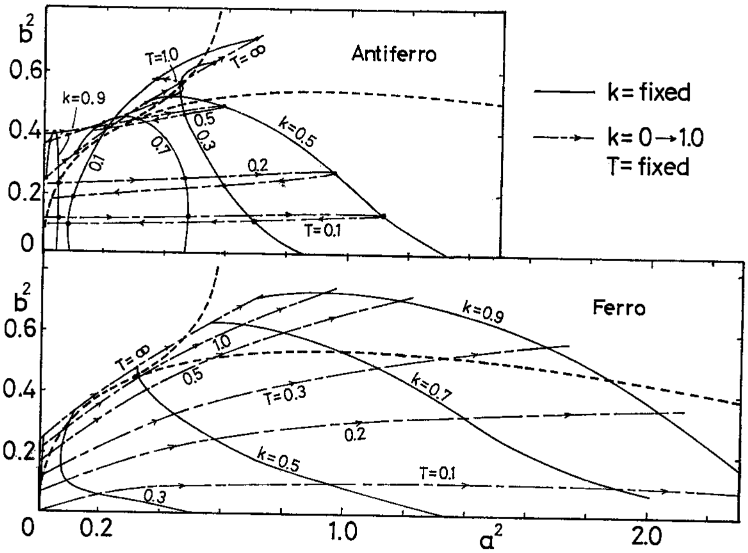



Fig. 9. Contours of constants $k$ and $T$ in unit $2JS^2/k_B$ (classical chain).

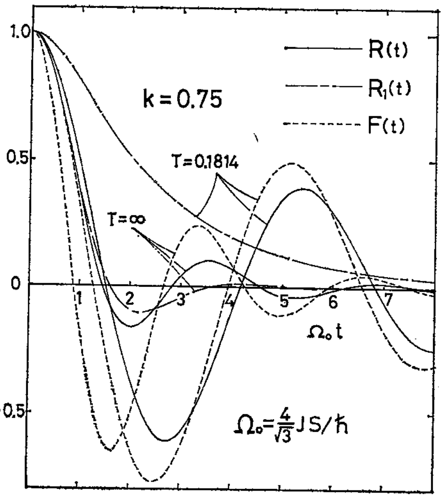



Fig. 10. Time correlations $R_i(t)$ (classical antiferromagnetic chain).

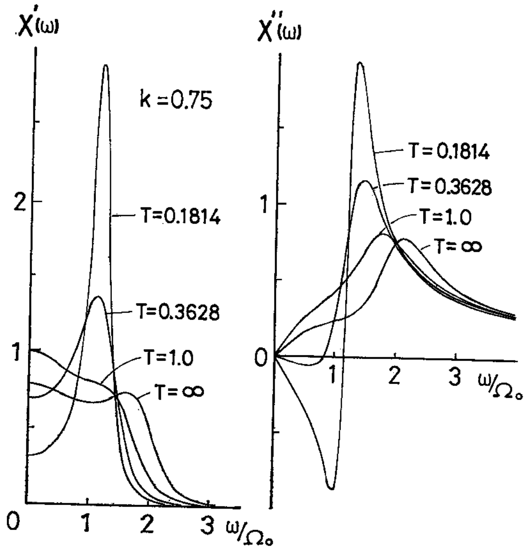



Fig. 11. Canonical correlation spectra $X(\omega)=X'(\omega)-X''(\omega)$ (classical antiferromagnetic chain).

The canonical correlation function, their Laplace transforms and the Cole-Cole plot are shown in Figs. 10, 11 and 12, respectively, for a fixed $k$ ($k=0.75$) and for several temperatures.

The situation $T\to\infty$ is to be compared with the published theoretical works of Morita,[12] McLean and Blume[13] and with the computer simulation by Windsor.[14] The canonical correlation function obtained in the present work for $T\to\infty$ is shown in Fig. 13, together with the results obtained by the above authors. It is clear that our results practically coincide with the computer simlation due to Windsor, and the results of McLean and Blume are not far from ours.

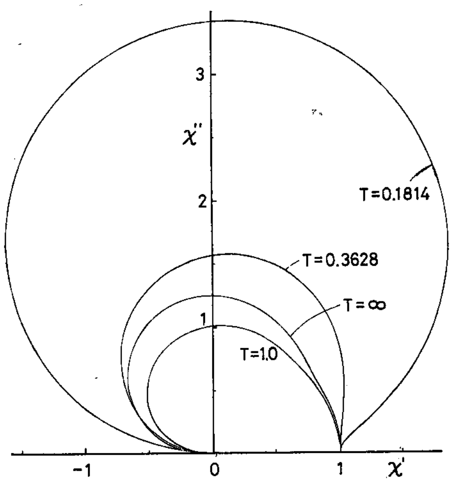



Fig. 12. The Cole-Cole plot (corresponding to Fig. 11).

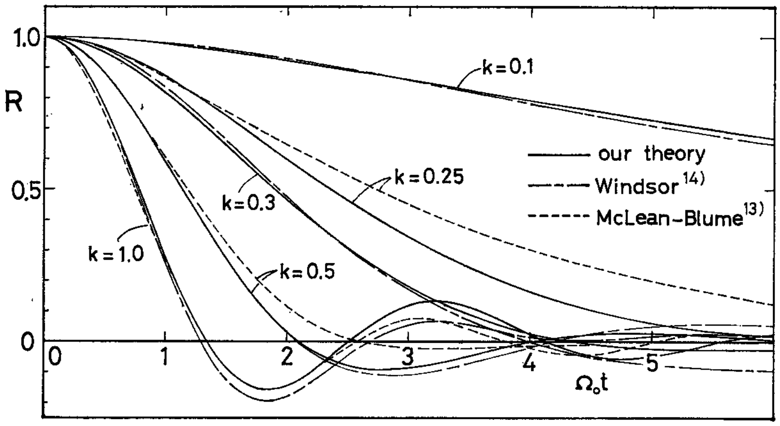



Fig. 13. Canonical correlations $R(k, t)$ of linear chain at $T\to\infty$.

For lower temperature situations there is a paper by McLean and Blume.[13] They calculated the canonical correlation under a provisional assumption on the coherence range in a low temperature paramagnet and found results compatible with TMMC data. Our results are also in good agreement with experiments and fairly close to their results. In the present treatment, however, no such assumption as theirs has been introduced, except that the spin system has been treated classically. Although the classical treatment might not be adequate in the lowest temperature limit, our approach seems more basic and may serve to check the validity of their assumption.

The actual comparison[2] with experimental data was made only in the case of TMMC,[11] i.e., an antiferromagnetic chain, the results of which are shown in Fig. 4 of Ref. 2). The agreement with the observation is satisfactory, considering that it covers the entire temperature range and the entire $k$ value range. The discrepancy in the lowest temperature range is probably due to the quantum fluctuation, which has been neglected in the theory.

In the case of $RbMnF_3$,[3)~5)] a three-dimensional antiferromagnet by which the present work was motivated, one has only approximate estimates of the second and fourth moments.[1)] Suppose the estimation is reliable, the only thing one can do is to guess the value of the sixth moment in such a way that the result fits the observed data. The results of the trial are shown in Fig. 14, where $\Delta_1^2$ and $\Delta_2^2$ are taken from the results of our calculation in the previous paper, and $\Delta_3^2$ is treated as a parameter. The quantitative fit one could actually attain is not quite satisfactory, concerning in particular the position of side peaks near the critical temperature.

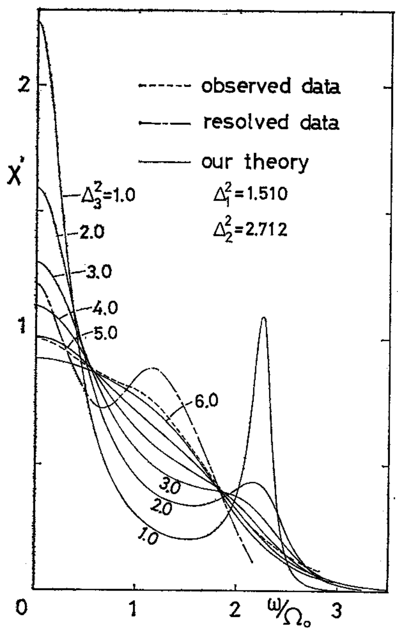



Fig. 14. Theoretical trial for $RbMnF_3$. (Predictions are closer to the data without resolution correction.)

The origin of the difficulty may be traced in two ways.

i) There is a possibility of overestimation of the fourth moment due to the decoupling approximation.[15)] The fit could have been better, provided the overestimation is of the order of 25%. The best fit is obtained for the situation in which $\Delta_1^2 \simeq \Delta_2^2 < \Delta_3^2$, which happens to be the most delicate case in so far as the line shape is concerned.

ii) The Gaussian approximation in third-order description might not be enough in the neighbourhood of the transition, because in this particular situation the higher-order correlation functions are not really well behaving.

## § 6. Discussion

In this section some discussions will be given on the plausibility of third-order description adopted in this paper. Obviously the most direct check will come from the knowledge of moments higher than the sixth.

Kwon and Gersch[16)] proposed a direct estimation of spin correlation function at a finite temperature on the basis of the cluster model. Although moments of any order may be easily calculated from their results, its dependence on wave number and temperature is not close to ours. In view of the nature of approximation they adopted, its applicability seems confined to a region corresponding to large $k$ values and to temperatures not very close to the critical point.

An analysis of more realistic model is obviously needed. In this context a computation of the eighth moment was made in the case of linear Heisenberg magnet,[17] and the results were found not to be inconsistent with Gaussian approximation at the third stage. This can be seen from Fig. 15.

Although it is not easy to obtain higher-order moments in general, the following observations of the behaviour of moments can be made in the temperature range which is essentially paramagnetic $(T > T_M \equiv \{2zJS(S+1)\}/3k_B)$ as a function of its order $n$, temperature and the wave number of the mode under consideration.

i) The characteristic behaviour on the macroscopic level, e.g., slowing-down phenomena either of kinematic or of thermodynamic origin, appears mainly in the lower moments and the higher order moments are relatively insensitive to such macroscopic character.

ii) The higher the order is, the less sensitive is the quantity $\Delta_n^2$ both to temperature and to the wave number.

As the absolute magnitude of the moment should be monotonically increasing with increasing order $n$, the above statement may asymptotically be visualized in the following way:

A) The asymptotic increase in the quantity $\Delta_n$ is expected to be normal, i.e.,

$$\Delta_n^2 \sim n . \quad (n \to \infty)$$

B) The quantity

$$\Delta r_n = (\Delta_{n+1}^2/\Delta_n^2) - (n+1)/n ,$$

which is chosen as a measure of deviation from the normality, is large and sensitive to the quantity observed only in the case of lower order moments. In other words $\Delta r_n$ converges to zero for all values of wave number and temperature, provided $n$ becomes large. These characteristics are born out in Fig. 15.

In the high-temperature limit, higher order $\Delta_i^2$ is definitely less sensitive to

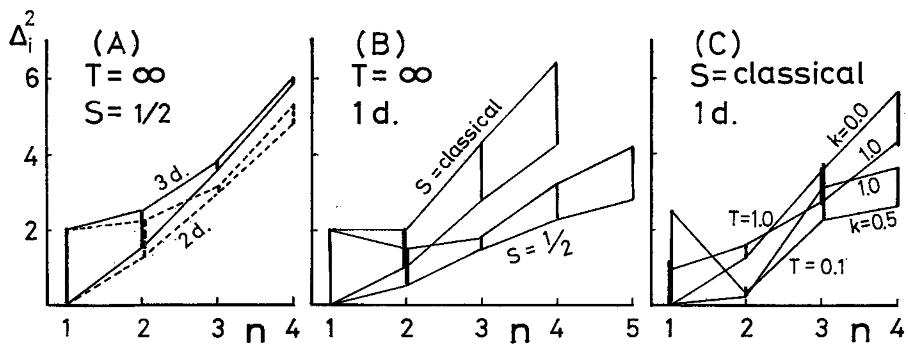



Fig. 15. The behaviour of $\Delta_n^2$.

the wave number as is recognized in $S=1/2$ spin system of which moments up to the tenth in order have been calculated by Morita,[18] as is shown in Fig. 16. It appears from Fig. 16 that $\Delta_{i+1}^2/\Delta_i^2$ depends little on $k$ for $i>3$ in two and three dimensions ($i>4$ in one dimension). This fact suggests strongly that one may replace the higher order correlation by a simple decreasing function, e.g., by Gaussian function. In this particular case an approximate asymptotic behaviour

$$\Delta_n^2 \sim n \quad (n\to\infty)$$

seems to exist.

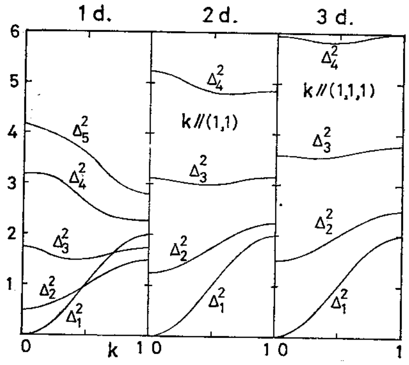



Fig. 16. Wave vector dependence of $\Delta_n^2$ ($S=\frac{1}{2}$, $T\to\infty$, cf. Ref. 18)).

Suppose the asymptotic behaviour $\Delta_n^2 \sim n$ for $n\to\infty$ is true, then the proper function one should take for $\overline{X}_2(\omega)$ is not a purely Gaussian function

$$\overline{X}_G(\omega)=\frac{1\,|}{|\,i\omega}+\frac{\Delta_3^2\,|}{|\,i\omega}+\frac{2\Delta_3^2\,|}{|\,i\omega}+\frac{3\Delta_3^2\,|}{|\,i\omega}+\cdots,$$

but a function

$$\overline{X}_{2G}(\omega)=\frac{1\,|}{|\,i\omega}+\frac{\Delta_3^2\,|}{|\,i\omega}+\frac{(4/3)\Delta_3^2\,|}{|\,i\omega}+\frac{(5/3)\Delta_3^2\,|}{|\,i\omega}+\cdots.$$

This is one theoretical possibility of improving the third-order description, and seems actually a better approximation if one looks into the result of stepping down in Fig. 3(b). That is, the Cole-Cole plot of $X_{2G}(\omega)$ appears much better than $X_G(\omega)$ as an approximation to $X_2(\omega)$.

In the low-temperature range, i.e., for $T<T_M$, the above statements have not been confirmed in general; however, the main difference in spectral line shape is expected to be induced again by the lower order moments in particular. Although there exists no reliable calculation of moments in the ferromagnetic phase in three dimension, the calculated results for one dimensional Heisenberg chain appear to support this conjecture. In Fig. 15(c) the existence of quasi-spin wave is essentially indicated by a conspicuous dip at order $n=2$ as compared with normal increase. Similar dips at higher orders, provided they exist, may lead to an additional wiggling in the spectrum. On the whole, however, the spectral characteristics are essentially determined by lower order moments. Namely, it has turned out that the inclusion of the knowledge of the eighth moment does not affect the position and width of the quasi-spin waves in any essential way as compared with those computed on the basis of moments up to the sixth. Moreover, the spectral phase diagram in Ref. 2) (Fig. 2) is qualitatively invariant under this modification.

Let us finally make several comments on the work hitherto published. Lovesey et al.[19] adopted an approach very similar to ours; however, they based their step-up process on the knowledge of the second and fourth moments only. Then they approximated $R_2(t)$ by a simple exponential function $e^{-t/\tau}$ with a provisional estimate of the rate constant $\tau^{-1}$ (or $\Delta_3$ in our terminology) based on the quantity $\Delta_2$. First, the exponential decay corresponds to a situation $\Delta_3 \ll \Delta_4$, which does not seem to be the case with this particular example. Secondly, the temperature dependences of $\Delta_2$ and $\Delta_3$ at low temperature are different from each other in the case of one-dimensional spin system, as was seen in our previous paper,[2] which fact cast doubt on their procedure. In fact the width of the spin wave peak is proportional to $T^{3/2}$ in their case, in contrast to $T$ in our case. This point is to be checked by more detailed experiments.

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**Appendix**

*—Higher order correlations leading to the Gaussian spectrum—*

It is known that a continued fraction

$$X(\omega) = \frac{1}{i\omega}\Big/ + \frac{a^2}{i\omega}\Big/ + \frac{2a^2}{i\omega}\Big/ + \frac{3a^2}{i\omega}\Big/ + \cdots \qquad \text{(A1)}$$

corresponds to a complex Laplace transform of a Gaussian correlation function $R(t) = \exp(-a^2t^2/2)$. Suppose $X(\omega)$ corresponds to the polarization correlation, then one may step down to the residual torque correlation

$$X_1(\omega) = \frac{1}{i\omega}\Big/ + \frac{2a^2}{i\omega}\Big/ + \frac{3a^2}{i\omega}\Big/ + \cdots . \qquad \text{(A2)}$$

One may repeat the above process and successively step down to $X_n(\omega)$. This has been done numerically and the shapes of successive $X_n(\omega)$'s are plotted in Fig. 1.

The correlation spectrum of order $n$ is defined by

$$X_n(\omega) = \frac{1}{i\omega}\Big/ + \frac{(n+1)a^2}{i\omega}\Big/ + \frac{(n+2)a^2}{i\omega}\Big/ + \cdots . \qquad \text{(A3)}$$

It is clear that $\Delta_n^2 \sim n\,(n \gg 1)$, and the ratio of the first two parameters is given by $r_{n+1} = (n+2)/(n+1)$. In the limit $n \to \infty$, we get $r_n \to 1$, which means that

$$X_\infty(\omega) = \frac{1}{i\omega}\Big/ + \frac{a^{*2}}{i\omega}\Big/ + \frac{a^{*2}}{i\omega}\Big/ + \cdots = \frac{1}{i\omega + a^{*2}X_\infty(\omega)} . \qquad \text{(A4)}$$

Although the "scale" $a^*$ of the distribution is ever increasing, it is possible to discuss the *"shape"* of the asymptotic distribution. Equation (A4) may be solved directly, i.e.,

$$X_\infty(\omega) = X'_\infty(\omega) - iX''_\infty(\omega) = \frac{1}{2a^{*2}}(-i\omega \pm \sqrt{-\omega^2 + 4a^{*2}}) \ . \qquad \text{(A5)}$$

Using the relation (8) in the text, it is found that

$$\chi'_\infty(\omega) = 1 - \omega^2/2a^{*2}\,, \qquad \text{(A6)}$$

$$\chi''_\infty(\omega) = \omega\sqrt{4a^{*2} - \omega^2}/2a^{*2}\,. \qquad \text{(A7)}$$

From (A6) and (A7), one can easily verify the relation

$$[\chi'_\infty(\omega)]^2 + [\chi''_\infty(\omega)]^2 = 1\,. \qquad \text{(A8)}$$

This is a semi-circle $C$ shown in Fig. 1 with a centre at the origin and with radius unity, and the shape of the real and and imaginary parts is shown in Fig. 2 in a reduced scale with its time domain function $R_\infty(t) = J_1(2a^*t)/a^*t$, where $J_1(x)$ is the Bessel function of first order. This is, so to speak, a *"proto-Gaussian"* asymptotic distribution in so far as the line shape is concerned.

When there exists no appreciable kinematic difference between successive scaling parameters $\Delta_n^2$ appearing in the continued fraction, the magnitude of $\Delta_n^2$ will be determined solely by a single coupling parameter and the combinational statistics. This means that progression will be proportional with that of natural numbers. Although in a real physical system there often appears specific behaviour due to either kinematic or thermodynamic origin in the lower order moments, the nature of the higher order moments are expected to be normal in many cases.

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