Divergences in the Space-Time Correlation Functions for the Heisenberg Magnet in One Dimension*

J. F. Fernandez and H. A. Gersch
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332

(Received 4 March 1968)

Carboni and Richards have performed exact numerical calculations of the time-dependent two-spin correlation function $\langle S_1^z(t)S_1^z(0)\rangle$ as $T\to\infty$ for a finite one-dimensional Heisenberg system. The non-Gaussian character of their result was characterized by a steep rise near zero frequency for the Fourier transform. We show here that these characteristics result from the inclusion of a Lorentzian form for $S(k,\omega)$, the paramagnetic scattering function at small wave vectors. We also prove that $\langle S_1^z(t)S_1^z(0)\rangle_\omega$, the time Fourier transform of $\langle S_1^z(t)S_1^z(0)\rangle$ in the $T\to\infty$ limit, obeys the inequality $\langle S_1^z(t)S_1^z(0)\rangle_\omega \geq \text{const}\times \ln|1/\omega|$ as $\omega\to 0$ for a one-dimensional system. We discuss the probable divergence of the same quantity in two dimensions.

I. INTRODUCTION

INTERPRETATION of experimental investigations of magnetic resonance linewidths, and neutron inelastic magnetic scattering requires knowledge of the time dependence of the spin-spin correlation functions. Recently, Carboni and Richards[1] reported exact numerical calculations of the two-spin correlation functions $\langle S_1^z(t)S_1^z(0)\rangle$ for finite linear chains of spin-$\frac{1}{2}$ particles coupled by a nearest-neighbor Heisenberg exchange interaction. They used certain extrapolation procedures to predict results for the infinite one-dimensional system. The results for the frequency transform $\langle S_1^z(t)S_1^z(0)\rangle_\omega$ showed a clearly non-Gaussian form, contrary to the predictions of Kubo and Tomita[2] and, in particular, a very steep rise near zero frequency (see Fig. 1).

In this paper we do three things. First, we show qualitatively that one expects the steep rise obtained by Carboni and Richards at zero frequency, coming from the inclusion of a Lorentzian form at small wave vectors k in the paramagnetic scattering function $S(k,\omega)$ from which one obtains the function $\langle S_1^z(t)S_1^z(0)\rangle_\omega$ by integration over k. Next, we assume $S(k,\omega)$ to be a properly weighted superposition of modified Gaussian and Lorentzian forms yielding the correct infinite-temperature values for the second and fourth moments of ω. A subsequent numerical calculation of $\langle S_1^z(t)S_1^z(0)\rangle_\omega$ from this assumed form of $S(k,\omega)$ is shown to agree in general with results of Carboni and Richards. Finally, we prove $\langle S_1^z(t)S_1^z(0)\rangle_\omega \geqslant \text{const}\times\ln|1/\omega|$ for a one-dimensional anisotropic Heisenberg system.

II. SCATTERING FUNCTION AND CORRELATION FUNCTION

The frequency transform of the time correlation function $\langle S_1^z(t)S_1^z(0)\rangle_\omega$ is related to $S(k,\omega)$, the paramagnetic scattering function, in the following way. $S(k,\omega)$ is defined to be

$$S(k,\omega)=\sum_j e^{i\mathbf{k}\cdot\mathbf{j}}\langle \mathbf{S}_1(t)\cdot\mathbf{S}_j(0)\rangle_\omega, \qquad (1)$$

where

$$2\pi\langle \mathbf{S}_1(t)\cdot\mathbf{S}_j(0)\rangle_\omega=\int_{-\infty}^{\infty} e^{i\omega t}\langle \mathbf{S}_1(t)\cdot\mathbf{S}_j(0)\rangle dt. \qquad (2)$$

Above the Curie temperature, for the isotropic Heisenberg magnet, the x-x, y-y, and z-z components of the spin correlation functions are all equal, hence

$$S(k,\omega)=3\sum_j e^{i\mathbf{k}\cdot\mathbf{j}}\langle S_1^z(t)S_j^z(0)\rangle_\omega. \qquad (3)$$

Integration of S and k leaves only the autocorrelation function

$$\frac{v}{(2\pi)^n}\int S(k,\omega)d^nk=3\langle S_1^z(t)S_1^z(0)\rangle_\omega, \qquad (4)$$

where v is the volume of the unit cell and n is the dimensionality of the crystal, $n=1, 2, 3$. Equation (4) establishes the desired connection between the spin correlation function and the paramagnetic scattering function. de Gennes[3] and Marshall[4,5] have reported exact infinite-temperature values of the second ($m=2$) and fourth ($m=4$) moments for ω:

$$\langle\omega^m\rangle_k=\int_{-\infty}^{\infty}\omega^m S(k,\omega)d\omega \Big/ \int_{-\infty}^{\infty}S(k,\omega)d\omega. \qquad (5)$$

These values are, for $s=\frac{1}{2}$ and in units where $\hbar=1$,

$$\langle\omega^2\rangle_k=2J^2(\gamma_0-\gamma_k),$$
$$\langle\omega^4\rangle_k=4J^4(\gamma_0-\gamma_k)(\tfrac{7}{2}\gamma_0-\tfrac{3}{2}\gamma_k-3). \qquad (6)$$

Here $\gamma_k=\sum_\delta e^{ik\delta}$ is summed over nearest neighbors δ. For large values of k, the ratio $\langle\omega^4\rangle_k/\langle\omega^2\rangle_k^2$ is quite close to 3, the result for a Gaussian $S(k,\omega)$, so that in

* Supported in part by National Science Foundation Grant No. GP 5639.

[1] F. Carboni and P. M. Richards, J. Appl. Phys. **39**, 967 (1968); R. N. Rogers, F. Carboni, and P. M. Richards, Phys. Rev. Letters **19**, 1016 (1967).

[2] R. Kubo and K. Tomita, J. Phys. Soc. Japan **9**, 888 (1954).

[3] P. G. de Gennes, J. Phys. Chem. Solids **4**, 223 (1958).

[4] W. Marshall, U.S. Dept. Comm., Office Tech. Serv., P. B. Rept. NBS **273**, 135 (1965).

[5] M. F. Collins and W. Marshall, Proc. Phys. Soc. (London) **92**, 390 (1967).

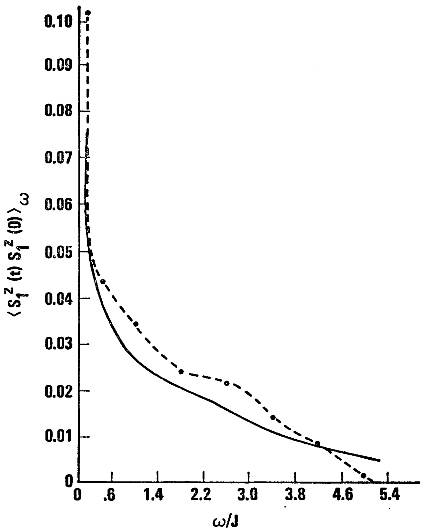

FIG. 1. The spin correlation function $\langle S_1{}^z(t)\, S_1{}^z(0)\rangle_\omega$ for a one-dimensional Heisenberg chain of $s=\frac{1}{2}$ spins at infinite temperatures. Dashed line is Carboni and Richards' results extrapolated to $N\to\infty$. Solid line is the combined result of Lorentzian and Gaussian forms, which are plotted separately in Fig. 2.

this large-k region S can be adequately expressed as a modified Gaussian.[5,6] The resulting $\langle S_1{}^z(t)\, S_1{}^z(0)\rangle_\omega$ will be Gaussian-like as a function of ω and quite different from the small-ω behavior shown in Fig. 1. However, for small values of k, the ratio $\langle\omega^4\rangle_k/\langle\omega^2\rangle_k{}^2$ differs drastically from the Gaussian value, as follows from Eq. (6), which predicts this ratio will go like $1/k^2$ for small k. In this region the Gaussian is replaced by a Lorentzian form,[3] which represents macroscopic spin diffusion:

$$\begin{aligned} S_L'(k,\omega) &= s(s+1)\,\Gamma_k/\pi(\omega^2+\Gamma_k{}^2), &\quad \omega\le\omega_c \\ &=0, & \omega>\omega_c \quad (7)\end{aligned}$$

where the choices

$$\Gamma_k=\pi\langle\omega^2\rangle_k{}^{3/2}/2[3\langle\omega^4\rangle_k]^{1/2}, \tag{8}$$

$$\omega_c=[3\langle\omega^4\rangle_k/\langle\omega^2\rangle_k]^{1/2} \tag{9}$$

are required to fit the two moments. From Eq. (6) we have the low-k behavior $\Gamma_k\sim k^2$, so that $S_L'(k,\omega)$ gets very large in the limit $\omega\to 0$ for small k, i.e., $S_L'(k,\omega)\sim 1/k^2$. Since the spin correlation function $\langle S_1{}^z(t)\, S_1{}^z(0)\rangle_\omega$ is obtained by integrating S over k, it follows that the spin correlation diverges in one and two dimensions as $\omega\to 0$, and must be the source of the steep rise in this function at zero frequency found by the numerical analysis of Carboni and Richards. Such a dimensional effect is perhaps not too surprising. The rate of macroscopic diffusion for the long times corresponding to small ω should be much slower in one- and two-dimensional lattices compared to three dimensions, since in the former the number of paths leading from a given spin site to far distant sites corresponding to small k are much reduced over the number of such paths in three dimensions.

[6] A numerical test of the adequacy of Gaussian and Lorentzian forms in the cluster model of the Heisenberg system has been carried out by T. H. Kwon and H. A. Gersch, Phys. Rev. **167**, 458 (1968).

To make a quantitative comparison of these ideas with the results of Carboni and Richards, we take $S(k,\omega)$ as a superposition of Gaussian and Lorentzian forms, with weighting functions chosen to make $S(k,\omega)$ essentially Gaussian at large k and Lorentzian at small k,

$$S(k,\omega)=S_L(k,\omega)+S_G(k,\omega), \tag{10}$$

where the Lorentzian form is given in terms of the previously defined $S_L'(k,\omega)$ as

$$S_L(k,\omega)=[(\gamma_0+\gamma_k)/2\gamma_0]S_L'(k,\omega). \tag{11}$$

The factor $(\gamma_0+\gamma_k)/2\gamma_0$ is largest at $k=0$ and goes to zero for k approaching the boundary of the Brillouin zone. For the one-dimensional $s=\frac{1}{2}$ case treated in Ref. 1, the Lorentzian width parameter Γ_k is given by

$$\Gamma_k=2^{1/2}J\pi(1-\cos ka)/(4-3\cos ka)^{1/2} \tag{12}$$

with a the lattice spacing. The Gaussian contribution $S_G(k,\omega)$ is taken to be

$$\begin{aligned} S_G(k,\omega) &= [(\gamma_0-\gamma_k)s(s+1)/2\gamma_0(2\pi\sigma^2)^{1/2}] \\ &\quad\times\exp(-\omega^2/2\sigma^2)[1+a_k(\omega^2/\sigma^2-1) \\ &\quad+b_k(\omega^4/\sigma^4-6\omega^2/\sigma^2+3)]. \quad (13)\end{aligned}$$

The factor $(\gamma_0-\gamma_k)/2\gamma_0$ is largest for $ka=\pi$, at the boundary of the Brillouin zone, and agreement with the exact second and fourth moments of ω is guaranteed by the choices

$$\begin{aligned} a_k &= \tfrac{1}{2}(\langle\omega^2\rangle_k/\sigma^2-1), \\ b_k &= \langle\omega^4\rangle_k/\langle\omega^2\rangle_k{}^2-3/24-\tfrac{1}{2}a_k. \quad (14)\end{aligned}$$

Equation (14) shows that the choice $\sigma^2=\langle\omega^2\rangle_k$ will yield $a_k=0$, and this is the usual choice. However, this choice complicates the integration over k of $S_G(k,\omega)$. A simpler procedure results from putting the parameter $\sigma^2=\langle\omega^2\rangle_{\max}=8J^2$, which occurs for $ka=\pi$. Then we approximate the fourth moment by its maximum value, achieved for $ka=\pi$, $\langle\omega^4\rangle_k=1.75\langle\omega^2\rangle_k{}^2$. Although this latter approximation becomes poor for small k, the contribution of this Gaussian part to the total $S(k,\omega)$ is also small in this region, and the approximation has the virtue of giving the integral of $S_G(k,\omega)$ over k in closed form. This integral yields, according to Eq. (4), the contribution to $\langle S_1{}^z(t)\, S_1{}^z(0)\rangle_\omega$. Written in terms of the variable $\omega'=\omega/J$, it is

$$\begin{aligned}\langle S_1{}^z(t)\, S_1{}^z(0)\rangle_\omega{}^G &= \frac{\exp(-\omega'^2/16)}{32\pi^{1/2}} \\ &\quad\times[1+\tfrac{1}{4}(\omega'^2/8-1)+0.028(\omega'^4/64-6\omega'^2/4)]. \quad (15)\end{aligned}$$

The dependence on ω' of this correlation function is shown in Fig. 2.

The corresponding contribution from $S_L(k,\omega)$, carried out by numerically integrating over k, is tabulated in Table I for the values ω' utilized by Carboni and Richards in their calculations. Figure 2 shows the Lorentzian contribution to $\langle S_1^z(t)S_1^z(0)\rangle_{\omega'}{}^L$. The total spin correlation function obtained by adding Gaussian and Lorentzian contributions is also depicted in Fig. 2 and is compared with Carboni and Richards's results in Fig. 1. Considering the arbitrariness both in our choice of the weighting factors $(\gamma_0 \pm \gamma_k)/2\gamma_0$ and in the choice of the parameters for the modified Gaussian, the agreement between our results and those of the extrapolated exact numerical calculations seems quite satisfactory. Certainly our results reproduce the sharp rise in the correlation function near zero ω.

III. PROOF OF DIVERGENCE OF $\langle S_1^z(t)S_1^z(0)\rangle$ IN ONE DIMENSION

We derive here a bound for $S^z(k,\omega)$ for a one-dimensional system with Hamiltonian given by

$$H=\sum_{i,j} J_{ij}(S_i^xS_j^x+S_i^yS_j^y)+\sum_{i,j} J_{ij}'S_i^zS_j^z \quad (16)$$

under the assumption that, in the $T\to\infty$ limit, $S^z(k,\omega)$ is a monotonically decreasing function of $|\omega|$ for small k. Notice that the Hamiltonian used here is more general than the one considered in the previous sections.

Equation (6) and the fact that $S^z(k,\omega)\geqslant 0$ yield the inequality

$$\int_{\omega_1}^{\omega_2} \omega^2 S^z(k,\omega)\,d\omega \leqslant ak^2, \quad (17)$$

where ω_1, ω_2 are any finite frequencies and a is a constant.

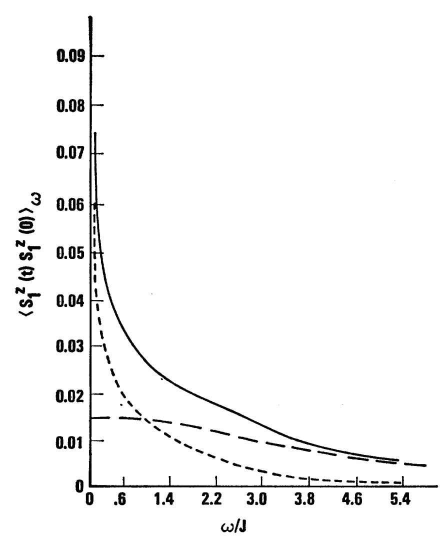

FIG. 2. Lorentzian and Gaussian contributions to the spin correlation function $\langle S_1^z(t)\,S_1^z(0)\rangle_\omega$ for a one-dimensional Heisenberg chain of $s=\frac{1}{2}$ spins at infinite temperature. Line of short dashes is the Lorentzian contribution as given in Table I. Line of long dashes is the Gaussian contribution as given by Eq. (15). The solid line represents their sum.

TABLE I. Values of the contribution of the Lorentzian paramagnetic scattering function $S_L(k,\omega)$ to the spin correlation function $\langle S_1^z(t)\,S_1^z(0)\rangle_{\omega'}{}^L$, for various values of reduced frequency, $\omega'=\omega/J$.

ω'	$\langle S_1^z(t)\,S_1^z(0)\rangle_{\omega'}{}^L$
0.1	0.0614
0.4	0.0257
1.0	0.0131
1.8	0.0071
2.6	0.0045
3.4	0.0031
4.2	0.0015
5.0	0.0007

The integral can be rewritten in terms of averages:

$$\langle\omega^2\rangle(\omega_1,\omega_2)\,\langle S^z(k,\omega)\rangle(\omega_1,\omega_2)\,(\omega_2-\omega_1)\leqslant ak^2, \quad (18)$$

where

$$\langle\omega^2\rangle(\omega_1,\omega_2)=\int_{\omega_1}^{\omega_2}\omega^2S^z(k,\omega)\,d\omega \Big/ \int_{\omega_1}^{\omega_2} S^z(k,\omega)\,d\omega \quad (19)$$

and

$$\langle S^z(k,\omega)\rangle(\omega_1,\omega_2)=\int_{\omega_1}^{\omega_2} S^z(k,\omega)\,d\omega \Big/ (\omega_2-\omega_1). \quad (20)$$

Now $\langle\omega^2\rangle(\omega_1,\omega_2)>\omega_1^2$ and $\langle S^z(k,\omega)\rangle(\omega_1,\omega_2)>S^z(k,\omega_2)$, as follows from the assumption that $S^z(k,\omega)$ is a decreasing function of $|\omega|$. Choosing $\omega_2=2\omega_1$, Eq. (18) becomes

$$\omega_1^3S^z(k,2\omega_1)\leqslant ak^2 \quad (21)$$

or

$$S^z(k,\omega)\leqslant bk^2/\omega^3, \quad (22)$$

where b is a constant. Next we utilize the sum rule

$$\int_0^\infty S^z(k,\omega)\,d\omega=\tfrac{1}{6}s(s+1)=c. \quad (23)$$

We break up the region of integration into two parts:

$$\int_0^{\omega_0} S^z(k,\omega)\,d\omega+\int_{\omega_0}^\infty S^z(k,\omega)\,d\omega=c, \quad (24)$$

where ω_0 is arbitrary. In the second integral on the left-hand side of Eq. (24) we use the inequality obtained in Eq. (22), and also the assumed monotonic character of S, to write

$$\omega_0S^z(k,\omega=0)+bk^2/2\omega_0^2\geqslant c. \quad (25)$$

The choice $\omega_0=c'k$ gives the strongest inequality:

$$S^z(k,\omega=0)\geqslant c/c'k-(b/2c'^3k). \quad (26)$$

We can choose c' large enough to make the right-hand side of Eq. (26) positive, so

$$S^z(k,\omega=0)\geqslant \alpha/k, \quad (27)$$

where α is some constant. The bound expressed by

Eq. (27) yields a weaker divergence for S^z than the Lorentzian form, which we previously showed leads to $S(k, \omega=0)\sim 1/k^2$. Our bound indicates a divergence in $\langle S_1^z(t)S_1^z(0)\rangle_\omega$ as $\omega\to 0$ for a one-dimensional Heisenberg system but not for a two-dimensional system. More physical information than the zeroth and second moments of ω are required to sharpen the inequality.

To establish the divergence of $\langle S_1^z(t)S_1^z(0)\rangle_\omega$ we now define a test function $S_0^z(k,\omega)$ which we suppose actually achieves the bound on $S^z(k,\omega)$ expressed by Eq. (27):

$$\lim_{\omega\to 0} S_0^z(k,\omega)=\alpha/k. \tag{28}$$

At the same time, $S_0^z(k,\omega)$ is required to have the same zeroth and second moments of ω as does $S^z(k,\omega)$, so that it also satisfies the inequality expressed by Eq. (22):

$$S_0^z(k,\omega)\leqslant bk^2/\omega^3. \tag{29}$$

For this test function, we establish the inequality

$$\int_0^{k_{\max}} S_0^z(k,\omega)dk \geqslant c_1 \ln|1/\omega|. \tag{30}$$

This is accomplished by utilizing the zeroth moment, or normalization condition, with the range of integration over ω subdivided,

$$\int_0^{\omega_1} S_0^z(k,\omega)d\omega+\int_{\omega_1}^{\omega_2} S_0^z(k,\omega)d\omega + \int_{\omega_2}^{\infty} S_0^z(k,\omega)d\omega = c, \tag{31}$$

with c a constant. Use of the inequality expressed by Eq. (29) in the third interval of integration gives the inequality

$$\omega_1 S_0^z(k,0)+S_0^z(k,\omega_1)(\omega_2-\omega_1)+bk^2/2\omega_2^2\geqslant c. \tag{32}$$

If we choose $\omega_2^2\gg bk^2$ and $\omega_1\ll k/\alpha$, then the first and third terms on the left side of Eq. (32) are numbers very small compared with unity, yielding

$$S_0^z(k,\omega)\geqslant d/k, \tag{33}$$

where d is a constant. Now integrate $S_0^z(k,\omega)$ over k:

$$\begin{aligned}\int_0^{k_{\max}} S_0^z(k,\omega)dk &\geqslant \int_{k_0}^{k_{\max}} S_0^z(k,\omega)dk \\ &\geqslant \int_{k_0}^{k_{\max}} (d/k)dk \\ &\geqslant d\ln(k_{\max}/k_0).\end{aligned} \tag{34}$$

We now choose the arbitrary lower limit $k_0=\omega c_1$, where $c_1\gg\alpha$. Then we have

$$\int_0^{k_{\max}} S_0^z(k,\omega)dk\geqslant \text{const}\times\ln|1/\omega|. \tag{35}$$

That the same inequality must hold for the paramagnetic scattering function $S^z(k,\omega)$ follows, since

$$\lim_{\omega\to 0} S^z(k,\omega)\geqslant \lim_{\omega\to 0} S_0^z(k,\omega) \tag{36}$$

and

$$\lim_{\omega\to 0}\int_0^{k_{\max}} S^z(k,\omega)dk\geqslant \lim_{\omega\to 0}\int_0^{k_{\max}} S_0^z(k,\omega)dk \geqslant \text{const}\times\ln|1/\omega|. \tag{37}$$

This establishes the logarithmic divergence of the z-z correlation function $\langle S_1^z(t)S_1^z(0)\rangle_\omega$. The proof holds equally well for the correlation function between different sites $\langle S_i^z(t)S_j^z(0)\rangle_\omega$ as can easily be shown.

The same proof can be used for the quantity $\langle\rho(x,t)\rho(x,0)\rangle_\omega$ for a one-dimensional, many-particle system independent of the form of the Hamiltonian. Here $\rho(x,t)$ is the particle-density operator. We would have to use, instead of Eq. (6), the sum rule for the second moment of $S(k,\omega)$ for this system, first derived by Yvon.[7]

Our rigorous inequality becomes an equality in the simple case of a free-particle system (with the appropriate constants inserted), as a simple calculation will show. Because of the analogy between the anisotropic Heisenberg magnet and the many-particle system,[8] we should expect this to be the case for the $J'=0$ model, the so-called x-y model. This is indeed the case, as Griffiths has shown.[9]

The bound we have derived does not predict a divergence of the correlation function in two dimensions. However, since the bound becomes the exact answer in the free-particle case, we are led to think that if we have collisions, i.e., $J'\neq 0$, the particles cannot move away from their initial positions so fast, [$\langle(x_i(t)-x_i(0))^2\rangle$ goes as t^2 for large t for free particles, whereas in the diffusion case it goes like t] and the diffusion equation should be applicable for long times, which implies a Lorentzian form for $S^z(k,\omega)$ and therefore a divergence in $\langle S_1^z(t)S_1^z(0)\rangle_\omega$ as $\omega\to 0$ in two dimensions also.

ACKNOWLEDGMENTS

We wish to thank Dr. R. B. Griffiths for a discussion which stimulated our interest in this subject. We also thank Dr. F. Carboni and Dr. P. M. Richards for sending us an unpublished report of their calculations.

[7] J. Yvon, Cahiers Phys. **14**, 1 (1943).
[8] P. R. Zilsel, Phys. Rev. Letters **15**, 476 (1965).
[9] R. B. Griffiths (private communication).