Progress of Theoretical Physics, Vol. 55, No. 1, January 1976

# Time Correlation Function in the One-Dimensional Heisenberg System at High Temperatures

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(Received May 19, 1975)

Moments in the two-time spin-pair correlation function are evaluated up to 16-th order for the one-dimensional Heisenberg system with S=1/2 at infinite temperature. The method of evaluations with a useful recurrence formula is reported. The current methods of treating the correlation function together with a newly proposed one are examined for both the spatially-Fourier-transformed correlation function and the auto-correlation function.

#### § 1. Introduction

Considerable attention has been paid to the spin dynamics in the Heisenberg system at high temperatures. In particular, the two-time spin-pair correlation function is the main subject. The spectral density function associated with the twotime spin-pair correlation function is closely related to the spectra of inelastically scattered neutrons<sup>10~71</sup> and to EPR line width.<sup>80,90</sup> Many theoretical studies of the two-time spin-pair correlation function have appeared. However there is no exact solution even for the one-dimensional lattice. The essential difficulty involved in our problem has clearly been described in Blume and Hubbard,<sup>100</sup> in which the references before 1969 have been extensively given.

Now, the moments associated with the spectral density function give us a valuable clue to improve the current methods and/or to examine their approximate nature. However the evaluation of moments becomes extremely difficult with increasing order. The moments have been evaluated up to 10-th order in the Heisenberg spin system for S=1/2 at infinite temperature,<sup>10,12)</sup> while the temperature-dependent moments up to 6-th order have been reported for the isotropic system with classical spins,<sup>4)</sup> all for one-dimensional case. We also mention the latest report on the 6-th moment for the isotropic system in any lattice with arbitrary spin.<sup>3)</sup>

It is one of our aims of this paper to present the moments up to 16-th order for the one-dimensional isotropic system with S=1/2 together with our method of evaluations. Another aim of this paper is to examine the approximate nature of some of the current methods of treating the two-time spin-pair correlation function, by using the obtained moments.

We shall particularly pay our attention to the method of continued fraction

expansion,<sup>13)</sup> which has widely been utilized. In this method some ingeneous approximation to the memory function is necessary for the cutoff of the continued fraction, as have been recongnized by various authours.<sup>3)~7)</sup> Among them Tomita and Mashiyama<sup>4)</sup> have analyzed the experimental spectrum of RbMnF<sub>3</sub> and have shown that the second-step memory function can be well approximated by a Gaussian function as an alternative of Lovesey and Meserve's exponential one.<sup>6)</sup> It is, however, desirable to look into the convergence of such methods by the use of the higher moments. In the present paper we also examine the other method, which will be called the approximate-stationary-memory-function method. This method seems implicit in Dupuis' paper.<sup>14)</sup>

In §2 we give the moments up to 16 th order for the one-dimensional isotropic system with S=1/2 together with a brief description of our method of evaluations. In §3 the results are given for the correlation spectra resulting from the Gaussian method, which is compared with those from the alternative one. In §4 we compute the auto-correlation function by the Gram-Charlier expansion, whose spectrum is compared with Carboni and Richards' result extrapolated from their calculation for finite chain system.<sup>80</sup>

#### § 2. Evaluation of moments

For the sake of simplicity we shall represent the Pauli spin operators  $(\sigma_{xj}, \sigma_{yj}, \sigma_{zj})$  at site j by  $(X_j, Y_j, Z_j)$ . The moment expansion of the two-time spin-pair correlation function at infinite temperature is merely the power series of it in time:

$$f(l,\tau) = \langle Z_i(0) Z_j(\tau) \rangle = \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n)!} M_{2n}(l) \tau^{2n}$$
(1)

with l=i-j. Here, by the use of Kubo's notation  $a^{\times}b=ab-ba$ , the 2*n*-th moment  $M_{2n}(j,i)$  is written as

$$M_{2n}(j,i) = \langle Z_i(H^{\times})^{2n} Z_j \rangle, \qquad (2)$$

In the reduced time  $\tau = Jt/\hbar$ , where our Hamiltonian is expressed in terms of the Pauli spin operators as

$$H = (1/2) \sum_{j} (X_{j} X_{j+1} + Y_{j} Y_{j+1} + Z_{j} Z_{j+1}).$$
(3)

As can be seen from the familiar expression

$$i^{-1}H^{\times}Z_{j} = Y_{j-1}Y_{j} - Y_{j}Z_{j+1} - X_{j-1}Y_{j} + X_{j}X_{j+1}, \qquad (4)$$

operation of  $(H^{\times})^n$  on  $Z_j$  generates various products of spin operators. A product of spin operators on k contiguous sites is called k-th order spin array. If the sequence of sites with spin operator is intermitted, we add 2 to the number of orders for each intervening site without spin operator. Thus for instance,  $X_j Y_{j+2}$ is a 4-th order array. Operation of  $i^{-1}H^{\times}$  on a k-th order spin array produces at most 4k spin arrays of the (k-1)-th and (k+1)-th orders. If two spin arrays coincide with each other by a translation, they are called dependent arrays. An assembly of dependent arrays is represented by the array with the leftmost spin operator at site 0. A spin array belonging to an assembly is generated from its representative by operating T(l) on the latter, making it translate by l sites. Independent spin arrays are denoted by  $\Pi_g$ , as numbered by the following way:  $\Pi_1 = Z_0$ ,  $\Pi_2 = Y_0 X_1$ ,  $\Pi_3 = X_0 Y_1$  and so on. The number of independent spin arrays appearing in  $(i^{-1}H^{\times})^n Z_0$ , which is denoted by  $h_n$ , increases with n approximately as  $h_n \sim 3^n$ .

Now the important point in our evaluation is the use of a recurrence formula. Let  $i^{-1}H^{\times}\Pi_g$  be

$$i^{-1}H^{\times}\Pi_{g} = \sum_{\hbar} \sum_{\delta} \sigma_{g\hbar}(\delta) T(\delta) \Pi_{\hbar} .$$
(5)

Here  $\delta$  runs over -1, 0, 1 and  $\sigma_{gh}(\delta) = 0, \pm 1$ , being determined from the evaluated result of commutators. Let us then introduce a many-dimensional vector  $A^{(n)}_{g,q}$ , whose *l*-th component is defined by

$$A^{(n)}{}_{g}(l) = i^{-n} \langle Z_{l}(H^{\times})^{n} \Pi_{g} \rangle.$$

$$\tag{6}$$

Inserting Eq. (5) into Eq. (6), we obtain the recurrence formula:

$$A^{(n)}{}_{g}(l) = \sum_{h} \sum_{\delta} \sigma_{gh}(\delta) A^{(n-1)}{}_{h}(l-\delta).$$
<sup>(7)</sup>

Since  $M_{2n}(j,i) = (-1)^n A^{(2n)}{}_1(j-i)$ , all independent spin-arrays appearing in  $(H^{\times})^m Z_0$  with  $m=0, 1, \dots, n$  are needed for the evaluation of  $M_{2n}(j-i)$ . If they are provided,  $A^{(2n)}{}_1$  can be obtained from Eq. (7) with the initial condition  $A^{(0)}{}_g(l) = \delta_{g1}\delta_{l0}$ . According to our procedure the number of independent coordinates necessary for the evaluation of  $A^{(2n)}{}_1$  is equal to  $(2n+1)h_n \sim 3^n n$ , owing to the recurrence formula. If one took the direct procedure with the use of Eq. (5), the corresponding number would be  $h_n^2 \sim 3^{2n}$  with a formidable figure.

The computed results for  $M_{2n}(l)$  are shown in Table I, whence  $M_{2n}(l)$  with l < 0 is obtained by  $M_{2n}(-l) = M_{2n}(l)$ . Another relation  $\sum_{l \ge 2} M_n(l) = 0$  as well as the above one checks our result. Besides these relations, some regularities are

Table I. Numerical values of  $M_{2n}(l)$ .

$l^{2n}$	0	2	4	6	8	10	12	14	16
0	1	4	44	652	11,636	242,816	5,896,200	166,988,876	5,522,663,076
1	i.	-2	-28	-456	-8,464	-177,424	-4,199,416		-3,410,887,248
2	İ		6	150	3,304	73,200	1,677,302	39,475,150	908,113,680
3		-		20	-728	-20,292	-525,316	-13,331,916	335,808,000
4		i			70	3,360	113,454	3,369,938	94,555,188
5	1		ł			-252	-15,048	597,168	-20,045,896
6	1						924	66,066	3,013,868
7		{				! :		-3,432	- 286,000
8	I I .		i			1			12,870

observed. We mention here  $M_{2n}(n) = (-1)^n (2n)!/(n!)^2$ .

The spatial Fourier transform of  $f(l, \tau)$  is given by

$$F(\kappa,\tau) = \langle Z(-\kappa,0)Z(\kappa,\tau) \rangle = \sum_{l} e^{i\kappa l} f(l,\tau), \qquad (8)$$

with

$$Z(\kappa) = N^{-1/2} \sum_{l} e^{i\kappa l} Z_l , \qquad (9)$$

whose spectral function  $\widetilde{G}(\kappa, \omega)$  is defined by

$$\widetilde{G}(\kappa,\omega) \equiv \int_{-\infty}^{\infty} d\tau \ e^{i\omega\tau} F(\kappa,\tau) \,. \tag{10}$$

The moments associated with  $\widetilde{G}(\kappa, \omega)$ ,  $M_{2n}(\kappa)$ , is merely a spatial Fourier transform of  $M_{2n}(l)$ :

$$M_{2n}(\kappa) = \sum_{l} M_{2n}(l) e^{i\kappa l} .$$
 (11)

 $M_{2n}(\kappa)$  can be written in terms of

$$X = 2(1 - \cos \kappa) \tag{12}$$

as follows:

$$M_{2n}(\kappa) = \sum_{m} M_{2n}^{m} X^{m} , \qquad (13)$$

where the coefficients  $M_{2n}^m$  obtained from Table I are tabulated in Table II.

The figures appearing along the diagonal line in the table are again given by  $(2n)!/(n!)^2$ . These are only non-vanishing coefficients which one would get for the XY-model.<sup>15</sup> Along the line contiguous to the diagonal one we have  $M_{2n}^{n-1} = [(2n)!/(n+1)!(n-2)!]$ . Otherwise there is no regularity as far as we could observe.

### § 3. Use of the continued fraction method

Let  $G(\kappa, s)$  be the Laplace transform of  $F(\kappa, \tau)$  as is defined by

2n160 2 4 6 8 10 1214 т 0 1 2 36 680 19,792 745,724 33,359,704 1,693,659,992 1 4  $\mathbf{2}$ 6 30 336 6,048 139,106 3,288,272 46,799,100 18,855,000 3 20 168 2,232 40,876 882,908 12,870 245,102 5,279,300  $\mathbf{4}$ 70 840 3,960 68,640 1,371,240 5 25218,018 6 924 348,348 7 3,432 80,080 12,870 8

Table II. Numerical values of  $M_{2n}^{m}$ .

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$$G(\kappa, s) = \int_0^\infty d\tau \ e^{-s\tau} F(\kappa, \tau), \quad s > 0.$$
<sup>(14)</sup>

This function may be represented by

$$G(\kappa, s) = 1/[s + \mathcal{A}_1^2(\kappa)G_1(\kappa, s)]$$
(15)

in accordance with Mori's treatment.<sup>13)</sup> Here  $G_1(\kappa, s)$  is the Laplace transform of the memory function which is called the first step memory function. The first step memory function is in turn expressed in terms of the second step memory function, both in their Laplace transforms, again in accordance with Mori's treatment. By this way one gets

$$G_{n-1}(\kappa, s) = 1/\lceil s + \mathcal{A}_n^2(\kappa) G_n(\kappa, s) \rceil,$$
(16)

where  $G_n(\kappa, s)$  is the Laplace transform of the *n*-th step memory function. The coefficients  $\mathcal{A}_n^{\ 2}(\kappa)$  are expressed in terms of the moments of  $F(\kappa, \tau)$ :<sup>13), 14)</sup>

$$\begin{aligned} & \mathcal{A}_{1}^{2}(\kappa) = M_{2}(\kappa), \\ & \mathcal{A}_{2}^{2}(\kappa) = M_{4}(\kappa) / M_{2}(\kappa) - M_{2}(\kappa), \\ & \mathcal{A}_{3}^{2}(\kappa) = [M_{6}(\kappa) / M_{2}(\kappa) - \{M_{4}(\kappa) / M_{2}(\kappa)\}^{2}] / \mathcal{A}_{2}^{2}(\kappa) \end{aligned}$$
(17)

and so on. Figure 1 shows the  $\mathcal{A}_n^2(\kappa)$ 's versus *n* for several values of  $\kappa$ .

Now the residual part, namely, the Laplace transform of the higher order memory function cannot be neglected as is widely known. If the n-th step memory function converges to a limiting function with increase of n, the approximate limiting memory function may be obtained if one puts

$$G_{n+1}(\kappa, s) = G_n(\kappa, s).$$
(18)

The above equation leads to<sup>14)</sup>

$$G_n(\kappa, s) = \left[ -s + \{s^2 + 4\mathcal{I}_{n+1}^2\}^{1/2} \right] \times (2\mathcal{I}_{n+1}^2)^{-1}.$$
(19)

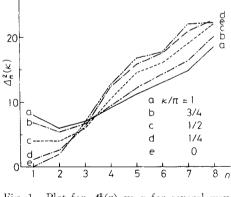


Fig. 1. Plot for  $\mathcal{A}_n^2(\kappa)$  vs *n* for several wave vectors.

However the existence of the limiting memory function is not necessarily required. If one could find a step number n at which  $\mathcal{A}_n^2$  is stationary with respect to the variation of n, the condition (18) would be valid there. In this sence the cutoff procedure with the use of Eq. (19) will be called the approximate-stationary-memory-function (ASMF) method, where  $G_n$  thus approximated will be referred to as the n-th ASMF.

In place of the above procedure, Tomita and Mashiyama<sup>5)</sup> assumed the second

step memory function to be Gaussian as given by

$$F_n(\kappa, \tau) = \exp(-\Delta_{n+1}^2 \tau^2/2), \quad (n=2)$$
(20)

which is compared with the inverse Laplace transform of Eq. (19), namely,  $J_1(2\Delta_{n+1}\tau)/(\Delta_{n+1}\tau)$  with  $J_1(x)$  the Bessel function of first order. The Laplace transform of Eq. (20) is written as

$$G_{n}(\kappa, -i\omega) = (\pi/2d_{n+1}^{2})^{1/2} \exp\left(-\omega^{2}/2d_{n+1}^{2}\right) \\ \times \left\{1 + 2i\pi^{-1/2} \int_{0}^{\omega/\sqrt{2}d_{n+1}} \exp\left(\tau^{2}\right) d\tau\right\}.$$
(21)

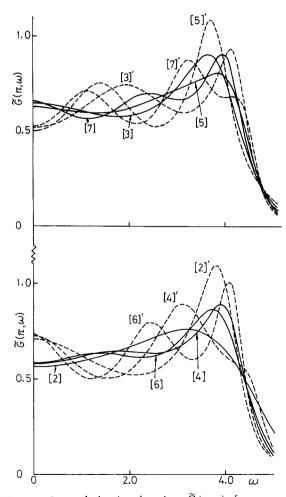


Fig. 2. Spectral density function  $\widetilde{G}(\kappa, \omega)$  for  $\kappa = \pi$ . The spectral density curve in the *n*-th ASMF is labeled by [n] and that in the *n*-th Gaussian approximation by [n]'. The angular frequency is reduced in units of  $\hbar/|J|$ .

We compute the spectral function for the spatial Fourier transform of the two-time spin-pair correlation function, which is given by  $\widetilde{G}(\kappa, \omega) = 2 \operatorname{Re} G(\kappa, -i\omega)$ . In Fig. 2 the computed results for  $\kappa = \pi$  are shown for both ASMF and Gaussian approximation with different step numbers of the cutoff, separately for even and odd step numbers. The area under the shown curves is automatically normalized to  $2\pi$ owing to the sum rule. Now, it appears that the Gaussian approximation gives peaks much sharper than ASMF as shown in Fig. 2. The curve [2] in the figure, where the cutoff has been done at the second step, is quite similar to that for classical spin in the second step Gaussian approximation,40 while it is not the case for the corresponding curve for spin one half in the latter approximation, [2]' in the same figure. It is to be noted that Lovesey and Meserve's exponential function<sup>6)</sup> gives us a sharper peak than the Gaussian approximation.

As can be seen in Fig. 2, ASMF seems more convergent than the Gaussian approximation. However the spectral curves in the higher approximation bring us another peak in the intermediate frequency region. We are not sure whether the new peak is spurious or not. Note that the fractional change in  $\Delta_n^2$  is the smallest between n=5 and n=6 as seen in Fig. This observation suggests that the 1. 4-th ASMF may be the most reasonable one. In Fig. 3 we show our computed results with the 4-th ASMF for several wave vectors and compare them with the previous ones. We note here that ASMF approximation gives results close to the Carboni-Richards spectral curves.

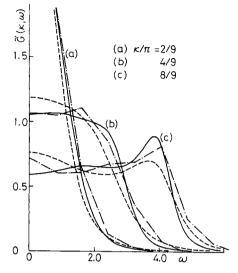


Fig. 3. Spectral density curves for several wave vectors. The angular frequency is reduced in units of ħ/|J|. Here the 4-th step ASMF: \_\_\_\_\_, the 2nd step Gaussian for classical spin:<sup>4)</sup> -----, Carboni and Richards' result: ----.

## §4. Use of the Gram expansion

The Gram expansion was applied first by Collins and Marshall,<sup>2)</sup> and later by various authors.<sup>16), 17)</sup> Here the starting function is a Gaussian one in accordance with the calculated second moment and

its modification is expanded in the derivatives of the same function where the expansion coefficients are fitted to the higher moments. The relevant expansion formula is obtained easily as given in Appendix A. The convergence of this method will be studied in detail particularly for the autocorrelation function.

The auto-correlation function  $f(0, \tau)$ defined by Eq. (1) with l=0, is in the best position for the purpose of studying the above-mentioned convergence, where one has the greatest number of available moments. There is also another point, as is described below. Let  $\mu_n$  be the ratio of  $M_{2n}(0)/(2n)!$  to  $M_{2n-2}(0)/(2n-2)!$ . Then a plot of  $\log[\log \mu_n + c]$  versus *n* is nearly a straight line with suitable choice

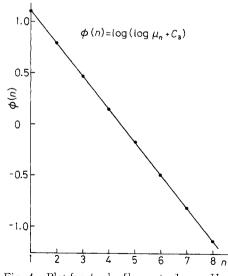


Fig. 4. Plot for  $\phi_n = \log[\log \mu_n + c_3]$  vs *n*. Here  $\mu_n = [M_{2n}(0)/(2n)!] + [M_{2n-2}(0)/(2n-2)!]$ and  $c_3 = 2.30373$ .

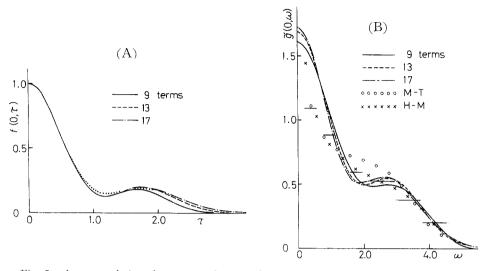


Fig. 5. Auto-correlation function and spectral density function in the Gram expansion. (A) Auto-correlation function  $f(0, \tau)$  as a function of the reduced time  $\tau = |J|t/\hbar$ . Carboni and Richards' extrapolated result is represented by the dotted lines. (B) Spectral density function  $\tilde{g}(0, \omega)$  as a function of the reduced angular frequency in units of  $\hbar/|J|$ . Carboni and Richards' result is represented by the horizontal lines. The open circles are McFadden and Tahir-Kheli's results<sup>20</sup> and the crosses Horiguchi and Morita's.<sup>21</sup>

of the constant c, as is manifested in Fig. 4. By this way we obtain the following 'empirical' formula:

$$M_{2n}(0) = (2n)! \exp[-c_1 \exp(-c_2 n) - c_3 n + c_4], \qquad (22)$$

with four parameters chosen to be  $c_1=11.0665$ ,  $c_2=0.320653$ ,  $c_3=2.30373$  and  $c_4=11.0411$ , using the least square fit. This formula can reproduce  $M_2$ ,  $M_4$ ,  $\cdots$ ,  $M_{16}$  to an accuracy of 0.02%. Thus we get some higher moments than  $M_{16}$ , as a result of the extrapolation with the aid of Eq. (22). (See also Appendix B).

The auto-correlation function  $f(0, \tau)$  thus obtained is plotted in (A) and the corresponding spectral density  $\tilde{g}(0, \omega)$  defined by the Fourier transform of  $f(0, \tau)$ , in (B), both of Fig. 5, where Carboni and Richards' result extrapolated from their finite chain calculations<sup>8</sup> is also shown. The convergence is notably slow and only three of our curves are plotted. The appearance of a broad maximum on the high frequency side remains unchanged even in the higher approximation with the use of extrapolated moments, as can be seen in the same figure. The hump thus found cannot clearly be seen in Carboni-Richards' horizontal lines, which are not in contradistinction to ours. The curve with 13 terms is almost the same as that with 17 terms except the center. The mentioned hump may be characteristic of the one-dimensional lattice with spin one half: Windsor's computer simulation result<sup>18</sup> shows a monotonically decreasing spectral curve, referring to the simple cubic lattice with classical spins.

We mention here the other kind of previous results for the auto-correlation function, basing its evaluation on De Leener and Résibois' integral equation.<sup>19)</sup> Here the relevant memory function is approximated by a Gaussian function and its modification may be treated by the same way as was done in the Gram expansion of the correlation function. The memory function approximated thus can be determined by the moments of the correlation function. The resulting spectral density functions have been published by McFadden and Tahir-Kheli<sup>20)</sup> and by Horiguchi and Morita<sup>21)</sup> respectively for the memory functions consisting of one and four terms. These results are shown also in Fig. 5(B).

The two-time spin correlation function for the nearest neighboring pair is another interesting subject. Unfortunately we could not succeed in finding any reliable extrapolation formula here. The single curve given in Fig. 6 is our result with the moments up to  $M_{16}(1)$  taken into account. Here our time correlation function  $f(1, \tau)$  is shown in (A) and its Fourier transform  $\tilde{g}(1, \omega)$  in (B), in comparison with Carboni and Richards' result.

In both Figs. 5(B) and 6(B), the spectral density near the center may not be reliable due to its singular behavior as pointed out by several authors.<sup>8), 22)</sup> (See also Månson<sup>23)</sup> for a recent discussion of the asymptotic behavior of the correlation function at long times.) The rise of the central peak with increase of the terms taken into account is clear in Fig. 6(B), where a bottom appears as a result of compensation of the area increase near the center.

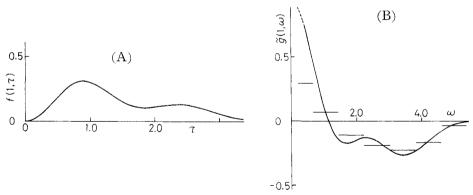


Fig. 6. Nearest neighbor correlation function  $f(1, \tau)$  and its spectral function  $\tilde{g}(1, \omega)$  in the Gram expansion. (A) The curve for  $f(1, \tau)$  with 8 terms in the expansion. (B) The curve for  $\tilde{g}(1, \omega)$  with the same approximation as in (A). The horizontal lines show the Carboni-Richards histgram.

### § 5. Conclusion and discussion

The moments associated with the two-time spin-pair correlation function have been evaluated up to the 16-th moment for the one-dimensional isotropic Heisenberg system with spin 1/2 at infinite temperature. The result is in agreement with the previous one up to the 10-th moment, of which the 6-th and 8-th moments had resulted from a direct count done by one of us (N.N.) without use of the computer, as quoted in Refs. 11) and 12). Owing to the reccurrence formula found in the direct count, the computation has been advanced to the higher moments as given in Table I.

Using the obtained moments, we have discussed the approximate nature of the continued fraction method with cutoff function particularly at the Brillouin zone boundary, which might be the worst case for the above method. Besides the Gaussian cutoff, another cutoff function called ASMF has been studied, resulting more convergent solution than the Gaussian one. However both the methods do not seem satisfactory as far as we could examine. It should be mentioned that the second step Gaussian cutoff has been suggested through an analysis of the experimental data for a three dimensional lattice. It has been pointed out that the 4-th ASMF seems more plausible than the higher ones for the considered system.

The Gram expansion method has been applied to the auto-correlation function with rather successful result. Due to a slow convergence of the expansion series we needed more moments than ours, which have been obtained with the help of an extrapolation formula. A hump in the relevant spectral curve has been predicted to exist, possibly as a characteristic of the one-dimensionality of our system.

#### Acknowledgements

We wish to thank Professor T. Nakamura for his continual encouragement and valuable discussions throughout the present study and Dr. J. Hama for his helpful discussions.

### Appendix A

### ----Gram's expansion-----

The Gram expansion will below be derived. The two-time correlation function for two spins separated by l sites,  $f(l, \tau)$ , is given by Eq. (1) of the text. For n < l, the  $M_{2n}(l)$ 's vanish identically as seen in Table II of the text. Then we put the series of  $f(l, \tau)$  as follows:

$$f(l,\tau) = \frac{(-)^{l}}{(2l)!} M_{2l}(l) \tau^{2l} \bigg[ 1 - \frac{1}{(2l+1)(2l+2)} \cdot \frac{M_{2l+2}(l)}{M_{2l}(l)} \cdot \tau^{2} + \cdots \bigg], \quad (A \cdot 1)$$

which may be written as

$$f(l,\tau) = \frac{(-)^{l}}{(2l)!} M_{2l}(l) \tau^{2l} \exp\left[-\frac{1}{2} (\omega_{l}\tau)^{2}\right] \sum_{s=0}^{\infty} \frac{1}{(2s)!} \cdot \frac{B_{2s}}{\omega_{l}^{2s}} (\omega_{l}\tau)^{2s} .$$
(A·2)

Here the width parameter  $\omega_i$  is chosen to be fitted to the initial decay and is given by Time Correlation Function in the One-Dimensional Heisenberg System 31

$$\omega_l^2 = \frac{2M_{2l+2}(l)}{(2l+1)(2l+2)M_{2l}(l)} . \tag{A.3}$$

The coefficient in Eq.  $(A \cdot 2)$  is then written as

$$B_{2s} = (2s)! \sum_{m=0}^{s} (-)^{m} \frac{(2l)!}{(2l+2m)!} \cdot \frac{M_{2l+2m}(l)}{M_{2l}(l)} \cdot \frac{1}{(s-m)!} \left(\frac{\omega_{l}^{2}}{2}\right)^{s-m}.$$
 (A·4)

The spectral density function is obtained as

$$\widetilde{g}(l,\omega) = \int_{-\infty}^{\infty} f(\tau) e^{i\omega\tau} d\tau$$

$$= \frac{\sqrt{2\pi}}{\omega_l} \cdot \frac{M_{2l}(l)}{(2l)!\omega_l^{2l}} \exp\left[-\frac{1}{2}\left(\frac{\omega}{\omega_l}\right)^2\right] \sum_{s=0}^{\infty} (-)^s \frac{1}{(2s)!} \cdot \frac{B_{2s}}{\omega_l^{2l}} H_{2l+2s}\left(\frac{\omega}{\omega_l}\right). \quad (A\cdot 5)$$

Here  $H_n(x)$  is the hermite polynomials. In Ref. 17), the width parameter  $\omega_l$  is assumed to be  $\omega_0$ , being independent of l.

### **Appendix B**

----Note on the extrapolated moments-----

After preparing the manuscripts the following paper drew our attention: A. Sur and I. J. Lowe, Phys. Rev. **B11** (1975), 1980. In this paper the moments associated with the auto-correlation function have been estimated by extrapolating the moments for the finite chain systems whose maximum number of spins is eleven. We shall below compare our extrapolated values of  $M_{2n}(0)$  with theirs for  $2n \ge 18$ , because both the values for 2n=16 are identical with each other up to the first five figures as it should be. We note here that their numerical values must perfectly be correct up to 2n=18. It should also be noted that their method of extrapolation is less reliable for  $M_{2n}(l)$  with larger l.

	$M_{2n}(0)$			
2n	ours	Sur-Lowe		
18	2.131×10 <sup>11</sup>	2.127×10 <sup>11</sup>		
20	$9.586 \times 10^{12}$	$9.480 \times 10^{12}$		
22 <sup>i</sup>	$5.002 \times 10^{14}$	$4.850 \times 10^{14}$		
24	$3.015 \times 10^{16}$	$2.821 \times 10^{16}$		
26	2.089×10 <sup>18</sup>	$1.844 \times 10^{18}$		
28	$1.653 \times 10^{20}$	$1.344 \times 10^{20}$		
30	1.49 ×10 <sup>22</sup>	$1.08 \times 10^{22}$		

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