Time-dependent correlations for spin Van der Waals systems

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The time-dependent autocorrelation function has been derived for the x component of the total spin for the $S = 1/2$ constant-coupling anisotropic Heisenberg model, i.e., the Van der Waals system. For $T > T_c$, the time correlation is shown to be Gaussian for both XY-like and Ising-like regimes of the model. For $T < T_c$, the correlation function is still Gaussian if the model is XY-like; but it is oscillatory if the model is Ising-like. Critical slow down appears only with the Ising-like system for this time-correlation function.

I. INTRODUCTION

By a spin Van der Waals system, we mean a system of lattice spins each of which is coupled to *all other* spins with *equal strength*. Such a system may be thought of as the high-density limit of an anisotropic Heisenberg model. Van der Waals systems belong to the class of mean-field models and are analytically soluble. Although limited in scope, the exact solutions for this class of systems can be useful in providing an insight into more complex systems. Recently, these mean-field models have taken on an added significance through the discovery that certain metal hydrides exhibit mean-field critical behavior.[1] The static properties of spin Van der Waals systems have been extensively studied by Kittel and Shore,[2] Niemeijer,[3] and Vertogen and DeVries.[4] They proved that equilibrium thermodynamic properties of spin Van der Waals systems are independent of spin dimensionality. That is, the isotropic Heisenberg and Ising systems in the high-density limit have identical equilibrium thermodynamic behavior.[5]

To our knowledge the dynamic properties of spin Van der Waals systems are not yet well known. It would be interesting to learn exactly how the Van der Waals systems respond to a time-dependent perturbation and dissipate it in time. It would also be. of interest to learn whether spin Van der Waals systems all have dynamically equivalent behavior, or whether they all belong to different dynamic universality classes.[6] For these purposes, we shall consider the $S = \frac{1}{2}$ constant-coupling anisotropic Heisenberg model and study the time-correlation function in appropriate physical regimes.[7] To obtain the time-correlation function, it is necessary to resolve the time-dependent part (by solving the Heisenberg equation of motion) and then to evaluate various quantities which do not depend on time. We shall study the transverse

component of the time-correlation function $S^{xx}(t)$ $= \langle S_x(t) S_x(0) \rangle$, where S_x is the x component of the total spin. Since S_x (or S_y) is not a constant of motion for the anisotropic Heisenberg model [see Eq. (2)], $S_x(t)$ has a nontrivial time evolution. On the other hand S_z is a constant of motion. Hence, the longitudinal component of the time-correlation function S^{zz} is trivial. It should be remembered that in this paper we discuss the *transverse*-correlation function only (whether the anisotropy is XY-like or Ising-like).[8]

For the one-dimensional nearest-neighbor XY model, the time-correlation function $S^{xx}(t)$ at the high-temperature limit is now known. Sur *et al.*,[9] Brandt and Jacoby,[10] and Capel and Perk[11] have shown that it is Gaussian. Since there is no phase transition in the one-dimensional nearest-neighbor XY model, the high-temperature form for the time-correlation function may be expected to persist, if in some modified way, into lower temperatures. We find that there seems to exist an interesting but evidently coincidental connection between this result for the one-dimensional nearest-neighbor XY model and our result for the constant-coupling XY model.

In Sec. II, our spin Van der Waals model is briefly described. In Sec. III, we review the technique of calculating the static quantities in the thermodynamic limit. In Sec. IV, the time-correlation function is derived for different physical regimes and also for different anisotropic limits. In Sec. V, we make a few concluding remarks.

II. DESCRIPTION OF THE MODEL

Consider a system of N $\frac{1}{2}$ spins, where N is considered to be a large number (the thermodynamic limit is always implied in our analysis). The components of the individual spin operator at site i are denoted by S_i^x, S_i^y, and S_i^z, $1 \leq i \leq N$, and those of

the total spin by S_x, S_y, and S_z, e.g.,

$$S_x = \sum_{i \neq 1}^{N} S_i^x .$$ (1)

In this system all spins are assumed to interact pairwise and with *equal* strength, independent of their relative distances. The interaction, however, is assumed to be anisotropic with respect to the spin components. With a conveniently chosen additive constant, the Hamiltonian of this system is given as follows:

$$\mathcal{H} = -\frac{1}{N} \sum_{i \neq j} \left[J(S_i^x S_j^x + S_i^y S_j^y) + J_z S_i^z S_j^z \right] - \frac{1}{4}(2J + J_z)$$

$$= -\frac{1}{N} (J\vec{S} \cdot \vec{S} - \lambda S_z^2) ,$$ (2)

where

$$\vec{S} = (S_x, S_y, S_z)$$ (3)

and

$$\lambda = J - J_z; \quad J > 0, \quad J_z > 0 .$$ (4)

We observe that the Hamiltonian is diagonal, i.e., with $\vec{S}^2 = S(S+1)$, $\mathcal{H} = f(S, S_z)$ only. Now λ can be either positive or negative. If $\lambda > 0$, the model is XY-like; and if $\lambda < 0$, it is Ising-like. We shall not consider the isotropic Heisenberg case ($\lambda = 0$) since all the components of the total spin, being conserved quantities, have trivial dynamic properties. The static properties of this system are not our main interest. But we shall begin by describing them first, since dynamic quantities must ultimately be expressed in terms of time-independent quantities.

III. STATIC PROPERTIES

The eigenstates of Hamiltonian (2) may be characterized by a set of quantum numbers $\{S$ and $S_z\}$. Evidently these states are multiply degenerate. The degeneracy must be accounted for in summing over states in evaluating, e.g., the partition function. For convenience we assume N to be an even number. The degeneracy g can be expressed as the difference between two binomial coefficients

$$g(S, S_z) = g(S) = \binom{N}{\frac{1}{2}N - S} - \binom{N}{\frac{1}{2}N - S - 1},$$ (5)

where $\frac{1}{2}N \geqslant S \geqslant 0$.[12] The above expression can be approximated for large N as

$$g(S) = 2^{N+1}[s/(1+s)]e^{-NW(s)} ,$$ (6)

where

$$s = 2S/N$$ (7)

and

$$W(s) = \frac{1}{2}[(1-s)\ln(1-s) + (1+s)\ln(1+s)]$$ (8a)

$$= 2(S/N)^2 + \frac{4}{3}(S/N)^4 + \cdots .$$ (8b)

It may be recognized that the degeneracy factor $g(S)$ represents the number of possible equivalent spin configurations (i.e., configurations having the same energy) for a given value of S.[13] Thus, $\ln g(S)$ is related to the entropy of the system,[14] and the free energy $F = \mathcal{H} - TS_e$ has a minimum at a certain value of the total spin S. The partition function may be evaluated in the following way:

$$Z = \text{Tr} e^{-\beta \mathcal{H}}$$

$$= \sum_{S=0}^{N/2} \sum_{S_z=-S}^{S} g(S)e^{\beta JS(S+1)/N - \beta \lambda S_z^2/N} .$$ (9)

Substituting for $g(S)$, we obtain

$$= 2^{N+1} \sum_{S} \left(\frac{2S/N}{1 + 2S/N} \right) e^{NG(S)} ,$$ (10)

where

$$G(S) = -W(S) + \frac{\beta JS(S+1)}{N^2} + \frac{1}{N}\ln \sum_{S_z} e^{-\beta \lambda S_z^2/N}$$ (11a)

$$\approx -(2 - \beta J)\left(\frac{S}{N}\right)^2 + \cdots + \frac{1}{N}\ln \sum_{S_z} e^{-\beta \lambda S_z^2/N} .$$ (11b)

We observe that $-\beta^{-1}G(S)$ is the free energy per spin for a fixed value of the *total* spin S. Evidently the phase factor $G(S)$ is the most important factor in determining the sum in Eq. (10). The dominant contributions come from the maximum of $G(S)$ given at some value of $S = S_0$. Hence, it will be found sufficient to evaluate the sum about S_0, where $G(S_0) = G_{\max}$.[15] Now S_0 must be a macroscopic number, i.e., $S_0 = f(N)$ in order that the prefactor of $e^{NG(S)}$ in the partition sum [the term inside the parentheses in Eq. (10)] does not vanish when evaluated about S_0. We shall find S_0 to be related to the long-range order of the system.

For both the XY-like and Ising-like systems there are two general classes which need be distinquished: high- and low-temperature regimes. For high temperatures, however, the XY and Ising systems are physically very similar so that they may be treated together. Our mathematical solutions for the partition function (given later in Sec. III) have a simple physical basis. This basis is first briefly discussed below and, we believe, will make the ensuing mathematical analysis rather transparent.

At high temperatures (i.e., $\beta J \ll 2$), the interaction energy $JS(S+1)/N^2$ is small compared with kT. Hence, the free energy or $-G$ is determined largely by the entropy term $-W$ [see Eq. (11a)]. At a *fixed* high temperature, the maximum entropy

(i.e., minimum F or maximum G) results from the state of N $\frac{1}{2}$ spins which are maximally *disordered*, i.e., $S_0 \neq O(N)$, so that $S_0/N \to 0$ as $N \to \infty$.[16] For both the XY and Ising systems we shall see that $S_0 = O(N^{1/2})$ confirming what is already known about the absence of long-range order at high temperatures.

At low temperatures (i.e., $\beta J \gg 2$), the interaction energy $JS(S+1)/N^2$ is now large compared with kT and it competes with the entropy in determining G. An *ordered* state will decrease the contribution of the entropy to G; but it can still increase G through the interaction energy, which of course increases with the ordering of spins. A maximum G can result from a such macroscopically ordered state of N $\frac{1}{2}$ spins, i.e., $S_0 = O(N)$ so that $S_0/N = O(1)$ as $N \to \infty$. This implies, of course, that there is a condensed phase for $\beta J \geq \beta_c J$ for some β_c, to be later determined.

A. High-temperature XY and Ising solutions

When $\beta = 1/kT$ is small, the phase factor of Eq. (10) is dominated by the entropy term $-W$. Since large values of S give little contribution to the partition function, we may approximate $W(S)$ by its first term in the series expansion, giving the following Gaussian form:

$$Z = 2^{N+1} \sum_{S=0}^{N/2} \left(\frac{2S}{N}\right) e^{-(2-\beta J)S^2/N}$$

$$\times \sum_{S_z=-S}^{S} e^{-\beta \lambda S_z^2/N}. \tag{12}$$

The maximum of the S summand of Eq. (12) is attained when $S = S_0 = O(N^{1/2})$. Thus, replacing the sums in Eq. (12) by integrals over $S = N^{1/2} x$ and $S_z = N^{1/2} z$, we have

$$Z = 2^{N+1} N^{1/2} \int_0^\infty dx\, e^{-(2-\beta J)x^2}$$

$$\times 2x \int_{-x}^x dz\, e^{-\beta \lambda z^2}, \tag{13}$$

where the upper limit on x has been extended to ∞ introducing little error. The above integral can be directly integrated by parts giving

$$Z = 2^{N+1} (\pi N)^{1/2} (2 - \beta J)^{-1} (2 - \beta J_z)^{-1/2}. \tag{14}$$

The leading correction to this expression comes from the neglected $(1+s)^{-1}$ [see Eq. (10)] and the correction will be a factor $N^{1/2}$ smaller than the above term in Eq. (14). The specific heat vanishes for T above T_c as it must for a mean-field model.[17]

Observe that our result [Eq. (14)] is independent of λ positive or negative. It must be remembered, however, that for the XY-like system the above result applied with $J > J_z$, whereas for the Ising-like system $J_z > J$. To show how the two different

conditions affect the result, we derive the fluctuations. For Van der Waals systems, the high-temperature spin fluctuations are given by[18]

$$Y^{xx} = \sum_{r=1}^N \langle S_0^x S_r^x \rangle = \frac{1}{2} \frac{\partial}{\beta \partial J} \ln Z, \tag{15a}$$

$$Y^{zz} = \sum_{r=1}^N \langle S_0^z S_r^z \rangle = \frac{\partial}{\beta \partial J_z} \ln Z. \tag{15b}$$

Thus, it follows directly from Eqs. (14) and (15) that, for the XY-like system, $Y^{xx} = \frac{1}{2}(\beta_c J - \beta J)^{-1}$ and $Y^{zz} = \frac{1}{2}(\beta_c J - \beta J_z)^{-1}$, and that for the Ising-like system, $Y^{xx} = \frac{1}{2}(\beta_c J_z - \beta J)^{-1}$ and $Y^{zz} = \frac{1}{2}(\beta_c J_z - \beta J_z)^{-1}$. That is, for the XY-like system, Y^{xx} is divergent but Y^{zz} is nondivergent at T_c; whereas, for the Ising-like system, the reverse holds. We have used here the results shown later in Sec. III that the XY-like system has $\beta_c J = 2$ and the Ising-like system has $\beta_c J_z = 2$.

B. Low-temperature XY solutions

For λ positive (i.e., XY regime), the S_z dependence in the partition function [see Eq. (12)] remains Gaussian for all β and the main contributions still come from $S_z = O(N^{1/2})$ or less. But for $\beta J < \beta_c J = 2$, the S dependence is not Gaussian, i.e., the previous high-temperature approximation for $W(S)$ is no longer valid. An examination shows that the phase factor $G(S)$ [see Eq. (10)] has a sharp maximum at $S = S_0 = O(N)$, so that $\exp N G(S)$ is sharply peaked at $S = S_0$. The condition $G'(S_0) = 0$ gives the familiar mean-field equation for the long-range order of the system,[13]

$$-W' + \beta J S_0/N = 0 \tag{16a}$$

or

$$S_0/N = \frac{1}{2} \tanh \beta J S_0/N. \tag{16b}$$

In the usual way, nontrivial solutions exist for T below $T_c = J/2K$ if $S_0 = O(N)$. Hence, defining $\sigma = 2S_0/N$, we may rewrite Eq. (16b) as

$$\sigma = \tanh(\beta/\beta_c)\sigma, \tag{16c}$$

with $\beta_c J = 2$.[19] Observe that Eq. (16c) is independent of J_z as long as λ is positive (i.e., $J > J_z$). That is, the critical temperature $\beta_c J = 2$ is universal for the Van der Waals XY-like system.

One can now expand $G(S)$ in Eq. (10) about S_0 and carry out the S integration by substituting $S = S_0 + Nx$. The S_z integration may be kept as before. Also since the two exponential functions are peaked at different regions with little overlap, the limits of integration in both cases may be extended. Thus we have

$$Z = g(S_0) e^{\beta J S_0^2/N} \int_{-\infty}^{\infty} dx \, e^{-\beta N B x^2} \int_{-\infty}^{\infty} dz \, e^{-\beta \lambda z^2}$$

$$= g(\tfrac{1}{2} N\sigma) e^{\beta J N \sigma^2/4} N\pi (\beta \lambda B)^{-1/2} [1 + O(N^{-1})] ,$$

(17a)

where

$$B = [2/(1 - \sigma^2)] - \beta J \approx (\beta - \beta_c) J \quad \text{as} \quad \beta \to \beta_c + . \quad (17b)$$

The specific heat follows directly from Eq. (17a) giving the usual mean-field result, in particular, $C/N = 3k$ at $T = T_c$

To calculate the fluctuations in the long-range order, it is necessary to include the corrections of order N^{-1} in the partition function. The corrections may be obtained by retaining second-order terms in the expansion of the S integrand about S_0.[15] The partition function valid to the order N^{-1} is as follows:

$$Z = Z_0 [1 + Z_1 N^{-1} + O(N^{-2})] , \quad (18a)$$

where Z_0 denotes the partition function given by Eq. (17a) and

$$Z_1 = \frac{1}{2B} \left(\frac{(\beta J)^2}{2} + \frac{2\beta J}{\sigma(1+\sigma)} - \frac{4}{\sigma(1+\sigma)^2} \right)$$

$$- 1/B^2 \left(\frac{5 - 4\sigma + 3\sigma^2 + 2\beta J\sigma(1 - \sigma^2)}{(1 - \sigma^2)^3} \right)$$

$$+ \frac{20}{3B^3} \frac{\sigma}{(1 - \sigma^2)^4} . \quad (18b)$$

C. Low-temperature Ising solutions

For negative λ (i.e., $J_z > J$), the sign of the phase factor $-\lambda \beta S_z^2$ changes. Thus, the S_z summation [see Eq. (10)] becomes very strongly dependent on its limits $(-S, S)$ so that, unlike in the XY system, it can influence the value of S_0 (the maximum of the phase factor $G(S)$). Introducing $\lambda = -\lambda'$, where λ' is a positive quantity, we express the S_z sum or integral in terms of Dawson's integral

$$\int_{-s}^{s} dS_z \, e^{\beta \lambda' S_z^2/N} = 2 e^{\beta \lambda' S^2/N}$$

$$\times \left(\frac{N}{\beta \lambda'} \right)^{1/2} D\left[S\left(\frac{\beta \lambda'}{N} \right)^{1/2} \right] , \quad (19)$$

where Dawson's integral is defined as[20]

$$D(y) = e^{-y^2} \int_0^y du \, e^{u^2} , \quad (20)$$

which is related to erfi y. We note that the argument of Dawson's integral $y = (\beta \lambda'/N)^{1/2} S$ can be large if $S = O(N)$. Since it is sufficient to expand $G(S)$ about $S = S_0$, one may thus replace $D(y)$ by its

asymptotic form [assuming of course that $S_0 = O(N)$]. The asymptotic expansion of $D(Y)$ is as follows:

$$D(y) = \sum_{n=0}^{\infty} \frac{2n!}{n!} (2y)^{-2n-1} = \frac{1}{2y} + \frac{1}{4y3} + \cdots \quad (21)$$

for y large.[20] Hence, we obtain for the phase factor $G(S)$,

$$G(S) = -W + \beta J(S/N)^2 + \beta \lambda'(S/N)^2$$

$$= -W + \beta J_z (S/N)^2 , \quad (22)$$

which is the same expression as that for the XY system with J_z replacing J. The condition that $G'(S_0) = 0$ gives the same mean-field equation as Eq. (16c), with J_z replacing J,

$$\sigma = \tanh(\beta/\beta_c)\sigma \quad (23)$$

but now with $\beta_c J_z = 2$. Thus, if $J_z > J$, the mean-field Eq. (23) is independent of J. That is, all Van der Waals Ising-like systems have the same value for T_c.[21]

The evaluation of the partition function follows the same procedure as that for the low-temperature XY system and we obtain,

$$Z = g(\tfrac{1}{2} N\sigma) e^{\beta J_z N \sigma^2/4} \frac{2N}{\beta \lambda' \sigma} \int_{-\infty}^{\infty} dx \, e^{-N B_z x^2}$$

$$= g(\tfrac{1}{2} N\sigma) e^{\beta J_z N \sigma^2/4} \frac{2}{\beta \lambda' \sigma}$$

$$\times \left(\frac{N\pi}{B_z} \right)^{1/2} [1 + O(N^{-1})] , \quad (24a)$$

where

$$B_z = 2/(1 - \sigma) - \beta J_z . \quad (24b)$$

If the correction terms are included, the partition function is

$$Z = Z_0 [1 + Z_1 N^{-1} + O(N^{-2})] , \quad (25a)$$

where Z_0 is given by Eq. (24a) and we have

$$Z_1 = \frac{1}{2B_z} \left(\frac{(\beta J)^2}{2} - \frac{2\beta J}{1+\sigma} + \frac{4}{(1+\sigma)^2} \right) - \frac{1}{B_z^2}$$

$$\times \left(\frac{1 - 4\sigma + 7\sigma^2}{(1 - \sigma^2)^3} + \frac{2\beta J\sigma}{(1 - \sigma^2)^2} \right)$$

$$+ \frac{2}{\beta \lambda' \sigma^2} + \frac{20}{3B_z^3} \frac{\sigma}{(1 - \sigma^2)^4} . \quad (25b)$$

All standard static quantities of interest can be readily obtained from our results for the partition function. In deriving the time-correlation function, it will be found necessary to evaluate still other static correlation functions such as $\langle S_z^{2n} \rangle$ and $\langle S_z^{2n} S_x^2 \rangle$ where n is any integer. These shall be treated at appropriate places in Sec. IV.

IV. TIME-CORRELATION FUNCTIONS

For the Van der Waals Hamiltonian Eq. (2), the time evolution of the total spin $\vec{S}$ may be obtained from the following set of Heisenberg equations of motion:

$$\dot{S}_x = -\omega_N(2S_zS_y + iS_x) , \qquad (26a)$$

$$\dot{S}_y = \omega_N(2S_zS_x - iS_y) , \qquad (26b)$$

$$\dot{S}_z = 0 , \qquad (26c)$$

where $\omega_N = \lambda/\hbar N$. These equations may be combined to give second-order differential equations separately for S_x and S_y. They are then readily solved to give[22]

$$S_x(t) = [\cos(2\omega_N t S_z)S_x - \sin(2\omega_N t S_z)S_y]e^{-i\omega_N t} , \qquad (27a)$$

$$S_y(t) = [\cos(2\omega_N t S_z)S_y + \sin(2\omega_N t S_z)S_x]e^{-i\omega_N t} , \qquad (27b)$$

$$S_z(t) = S_z , \qquad (27c)$$

where $S_x = S_x(t=0)$, etc.

The time evolutions of S_x and S_y appear to be oscillatory. In fact, however, they are rather complex owing to the presence of S_z, which is a constant in time but still an operator. Thus one must maintain the order of these noncommuting operators. Using the above results, we shall deduce the time-correlation function $\langle S_x(t)S_x(0)\rangle$ for the XY-like and Ising-like systems and study the time-dependent behavior for T above and below T_c for each of these systems. This time-correlation function is particularly useful since one can obtain from it the dynamic form factor or spectral density function, hence all other important dynamic quantities such as the relaxation function and dynamic susceptibility.[23]

For convenience we shall absorb the trivial exponential factor which appears in Eq. (27) in the definition of our time-correlation function by defining it as

$$\mathcal{S}^{xx}(t) = e^{i\omega_N t}\langle S_x(t)S_x(0)\rangle , \qquad (28)$$

which satisfies time-reversal symmetry. Using our results given above we obtain

$$\mathcal{S}^{xx}(t) = \langle \cos(2\omega_N t S_z)S_x^2 - \sin(2\omega_N t S_z)S_yS_x\rangle . \qquad (29)$$

Now since S_z is diagonal with respect to the eigenvectors of the Hamiltonian, we observe that, in the second term in Eq. (29), only the diagonal part of S_yS_x can contribute in the ensemble average. Hence, we can reduce Eq. (29) to the following:

$$\mathcal{S}^{xx}(t) = \langle \cos(2\omega_N t S_z)S_x^2\rangle + \tfrac{1}{2}i\langle\sin(2\omega_N t S_z)S_z\rangle , \qquad (30a)$$

which by expanding the trigonometric functions leads to

$$= \sum_{n=0}^{\infty} (-)^n \frac{(2\omega_N t)^{2n}}{2n!} \langle S_z^{2n}S_x^2\rangle$$

$$+ \tfrac{1}{2}i \sum_{n=0}^{\infty} (-)^n \frac{(2\omega_N t)^{2n+1}}{(2n+1)!}\langle S_z^{2n+2}\rangle . \qquad (30b)$$

Taking advantage of the XY symmetry, i.e.,

$$\langle S_z^{2n}S_x^2\rangle = \tfrac{1}{2}(\langle S_z^{2n}S^2\rangle - \langle S_z^{2n+2}\rangle) , \qquad (31)$$

we finally obtain

$$2\mathcal{S}^{xx}(t) = \sum_{n=0}^{\infty} (-)^n \frac{(2\omega_N t)^{2n}}{2n!} \langle S_z^{2n}S^2 - S_z^{2n+2}\rangle$$

$$+ i\sum_{n=0}^{\infty} (-)^n \frac{(2\omega_N t)^{2n+1}}{(2n+1)!} \langle S_z^{2n+2}\rangle . \qquad (32)$$

Observe that the time part has now been completely resolved from time-independent-correlation functions. To obtain the time-correlation function $\mathcal{S}^{xx}(t)$, one needs to know the two equilibrium correlation functions $\langle S_z^{2n}S^2\rangle$ and $\langle S_z^{2n}\rangle$ only. These equilibrium quantities depend on λ and also on the temperature. They may be evaluated in the manner of the partition function shown in Sec. III. We shall consider them in three different regimes: high-temperature XY-like and Ising-like, low-temperature XY-like, and low-temperature Ising-like.

A. High-temperature XY and Ising models

We shall evaluate $\langle S_z^{2n}\rangle$ and $\langle S_z^{2n}S^2\rangle$ and also a few derivative terms of interest closely following the static analysis previously given (see Sec. III A). For any integer n (including $n=0$),

$$\langle S_z^{2n}\rangle = Z^{-1} \sum_{S} \sum_{S_z} g(S)S_z^{2n} e^{-\beta \mathcal{K}(S,S_z)}$$

$$= Z^{-1} 2^{N+1}N^{n+1/2} \int_0^{\infty} dx\, e^{-(2-\beta J)x^2}$$

$$\times 2x \int_{-x}^{x} dz\, z^{2n} e^{-\beta\lambda z^2}$$

$$= \frac{2n!}{n!}\left(\frac{N}{4(2-\beta J_z)}\right)^n . \qquad (33a)$$

Observe that

$$\langle S_z^2\rangle = \tfrac{1}{2}N/(2-\beta J_z) , \qquad (33b)$$

which for the pure XY model becomes independent of the temperature, $\langle S_z^2\rangle = \tfrac{1}{4}$ for $T > T_c$. Similarly we have

$$\langle S_z^{2n} S^2 \rangle = Z^{-1} 2^{N+1} N^{n+3/2} \int_0^\infty dx\, e^{-(2-\beta J)x^2}$$

$$\times\, 2x^3 \int_{-x}^x dz\, e^{-\beta \lambda z^2} z^{2n}$$

$$= N\left(\frac{1}{2-\beta J} + \frac{1}{2}\frac{2n+1}{2-\beta J_z}\right)\frac{2n!}{n!}\left(\frac{N}{4(2-\beta J_z)}\right)^n .$$

(34a)

We note that by setting $n = 0$ in Eq. (34a) we have

$$\langle S^2 \rangle = \frac{1}{2}N[2/(2-\beta J) + 1/(2-\beta J_z)]$$ (34b)

and with Eq. (33b)

$$\langle S_x^2 \rangle = \langle S_y^2 \rangle = \frac{1}{2} N/(2-\beta J).$$ (34c)

The high-temperature spin fluctuations are all of order N and behave mean-field-like near T_c as was discussed in Sec. III. These may also be used to show that in the high-temperature regime the longitudinal and transverse factors of the static correlation functions decouple, e.g.,

$$\langle S_z^{2n} S_x^2 \rangle = \langle S_z^{2n}\rangle\langle S_x^2 \rangle .$$ (35)

Substituting Eq. (33a) and Eq. (34a) in Eq. (32), we obtain (after some simple sums)

$$S^{xx}(t) = [\langle S_x^2\rangle + i\tfrac{1}{2}\lambda t/(2-\beta J_z)\hbar]$$
$$\times\, e^{-(\lambda t)^2/(2-\beta J_z)\hbar^2 N}$$ (36a)

which evidently is valid for $0 \le t \lesssim t_{max}$, where $t_{max} = (\hbar^2 N/\lambda^2)^{1/2}$. Since $\langle S_x^2\rangle = O(N)$, for this range of time the time-correlation function valid to the leading order of N is given by

$$S^{xx}(t) \simeq \langle S_x^2\rangle\, e^{-(a_N t)^2},$$ (36b)

where

$$a_N(\lambda) = [\lambda^2/\hbar^2 N(2-\beta J_z)]^{1/2}.$$ (36c)

The time-correlation function satisfies time-reversal symmetry. It is Gaussian, implying that the dynamic form factor is also Gaussian.[18] Although our result [Eq. (36b)] applies equally to the XY-like and Ising-like systems (i.e., $\lambda > 0$ and $\lambda < 0$, respectively) the temperature-dependent behavior of the "width" of the Gaussian factor $a_N^{-1}(\lambda)$ is markedly different between the two systems. For the Ising-like system, the width narrows as the critical temperature is approached from above and the system exhibits critical slow down.

For the XY-like system, the situation is rather different. Here the static criticality is resolved from time, being contained in $\langle S_x^2\rangle$ and not in the width a_N^{-1}. Hence, the time-correlation function for the XY-like system cannot exhibit critical slow down.[18] For the pure XY model (i.e., $J_z = 0$ or $\lambda = J$), the width factor reduces to $a_N^{-1} = (\frac{1}{2}J^2/$

$\hbar^2 N)^{-1/2}$ which has no temperature dependence at all at high temperatures ($\beta_c J \lesssim 2$). This general form of the width, we believe, may be preserved if the number of interacting "near-neighbor" spins were reduced from N to a small number, say q. In this case we have for the width $a_q^{-1} = (\frac{1}{2}qJ^2/\hbar^2)^{-1/2}$, where we have restored the interaction strength from J/N to J. [Recall that N was introduced in Eq. (2) to keep the energy per spin finite in the limit $N \to \infty$.] It is interesting to note that a dynamic mean-field version of the three-dimensional nearest-neighbor (nn)XY model for $T > T_c$ gives exactly this form of the width for the time-correlation function, where q is the nn number.[18] Also, the same form is obtained for the time-correlation function for the one-dimensional nnXY model.[9-11]

B. Low-temperature XY model

We shall evaluate $\langle S_z^{2n}\rangle$ and $\langle S_z^{2n}S^2\rangle$ following the static analysis given in Sec. IIIB. For any integer n we have

$$\langle S_z^{2n}\rangle = Z^{-1} \sum_S \sum_{S_z} g(S) S_z^{2n} e^{-\beta \mathcal{H}(S, S_z)}$$

$$= \frac{2n!}{n!}\left(\frac{N}{4\beta\lambda}\right)^n .$$ (37)

Hence, $\langle S_z^2\rangle = N/2\beta\lambda$. For the pure XY model, it becomes $\langle S_z^2\rangle = N/2\beta J$, which at the critical point $\beta J = \beta_c J = 2$ assumes the high-temperature value, becoming independent of the temperature. In evaluating $\langle S_z^{2n}S^2\rangle$, we recall that the maximum contributions of S and S_z come from widely separated regions of N so that the two integrations have effectively two independent limits, i.e., the correlation function decouples,

$$\langle S_z^{2n}S^2\rangle = \langle S_z^{2n}\rangle\langle S^2\rangle .$$ (38)

We can directly evaluate $\langle S^2\rangle$ as, for example, in Eq. (37) giving

$$\langle S^2\rangle = (\tfrac{1}{2}N\sigma)^2 + \frac{N}{2B}\left(\beta J\sigma + \frac{3+\sigma}{1+\sigma}\right) - \frac{2N\sigma^2}{B^2(1-\sigma^2)^2},$$ (39)

where we have used σ and B previously defined (see Sec. IIIB). It follows that we have

$$\langle S_x^2\rangle = \frac{1}{2}\left[(\tfrac{1}{2}N\sigma)^2 + \frac{N}{2B}\left(\beta J\sigma + \frac{3+\sigma}{1+\sigma}\right) - \frac{2N\sigma^2}{B^2(1-\sigma^2)^2}\right] - \frac{N}{4\beta\lambda}.$$ (40)

Observe that $\langle S_x^2\rangle = O(N^2)$ unlike in the high-temperature case [see Eq. (34c)]. The second-order terms shown in Eq. (40) are needed when evaluating the spin fluctuation since $\langle S_x\rangle = O(N)$.

Using Eq. (37), Eq. (38), and Eq. (40) in Eq. (32), we obtain for the time-correlation function

$$S^{xx}(t) = \left(\langle S_x^2 \rangle + \frac{it}{2\hbar\beta} + \frac{t^2}{2\hbar^2\beta^2}\right) e^{-\lambda t^2/\hbar^2\beta N}, \quad (41a)$$

which evidently is valid for $0 \leq t \lesssim t'_{max}$, where $t'_{max} = (\hbar^2\beta N/\lambda)^{1/2}$. Hence, for this range of time, we may take the correlation function to be

$$S^{xx}(t) \simeq \langle S_x^2 \rangle e^{-(a'_N t)^2}, \quad (41b)$$

where $a'_N = (\lambda/\hbar^2\beta N)^{1/2}$. It is again Gaussian, but the width is now temperature dependent. As the temperature is lowered, the width grows as $T^{-1/2}$. As the temperature is raised, the width narrows to a finite value. For the pure XY model the width has no discontinuity at T_c, i.e., for $\lambda = J$, $a'_N = a_N$ at $\beta J = 2$. As in the high-temperature side, the static criticality is resolved from time [see Eq. (41b)] so that there cannot be critical slow down in this time-correlation function as $T \to T_c$.

C. Low-temperature Ising model

For λ negative, the evaluations of $\langle S_z^{2n} \rangle$ and $\langle S_z^{2n} S^2 \rangle$ must be proceeded with more care as pointed out in Sec. III C. Writing $\lambda = -\lambda'$ (where λ' is a positive quantity), we have for any integer n

$$\langle S_z^{2n} \rangle = Z^{-1} \sum_S g(S) e^{\beta JS^2/N}$$
$$\times \left(\sum_{S_z} S_z^{2n} e^{\beta\lambda'S_z^2/N} \right). \quad (42a)$$

The term inside the parentheses in Eq. (42a) may be handled as follows:

$$\sum_{S_z} S_z^{2n} e^{a S_z^2} = \left(\frac{\partial}{\partial a}\right)^n \sum_{S_z} e^{a S_z^2}$$
$$= \left(\frac{\partial}{\partial a}\right)^n [2a^{-1/2} e^{a S^2} D(a^{1/2}S)]$$
$$\simeq a^{-1} e^{a S^2} S^{2n-1} \left(1 - \frac{2n-1}{2aS^2} + \cdots \right), \quad (42b)$$

where $a = \beta\lambda'/N$, D is Dawson's integral [see Eq. (20)], and in obtaining Eq. (42b) the asymptotic expansion of Dawson's integral [Eq. (21)] has been used.

Substituting Eq. (42b) in Eq. (42a), we finally obtain

$$\langle S_z^{2n} \rangle = (\tfrac{1}{2}N\sigma)^{2n} \left[1 - \frac{2}{N\sigma} \left(2nA - \frac{n(2n-1)}{B_z\sigma}\right) + O(N^{-2}) \right], \quad (43a)$$

where

$$A = \frac{1}{B_z(1+\sigma)} + \frac{2\sigma}{B_z^2(1-\sigma^2)^2} - \frac{1}{\beta\lambda'\sigma} - \frac{\beta J}{2B_z}, \quad (43b)$$

and see Eq. (24b) for B_z.

Observe that for $n = 1$ we have

$$\langle S_z^2 \rangle = S_0^2[1 + O(N^{-1})],$$

where we recall that $S_0 = \tfrac{1}{2}N\sigma$. The terms of order N^{-1} in Eq. (43b) thus determine the spin fluctuations $Y^{zz} = \langle S_z S_z \rangle - \langle S_z \rangle^2$. We note that Y^{zz} are $O(N)$ and have critical behavior. In evaluating the time-correlation function $S^{xx}(t)$ with Eq. (43a), the terms of order N^{-1}, which also depend on n, will give additional time-dependent factors. To be consistent with the spirit of our expansion (i.e., an expansion in powers of N^{-1}), we must restrict largest values of n to be less than $O(N^{1/2})$.

Similarly, we obtain for any integer n,

$$\langle S_z^{2n} S_x^2 \rangle = S_0^{2n} N/2\beta\lambda'. \quad (44)$$

Observe that by setting $n = 0$ in Eq. (44), we get $\langle S_x^2 \rangle = O(N)$. It is linear in N, whereas the leading term of $\langle S_z^2 \rangle$ is quadratic in N. The spin fluctuations Y^{xx} have no anomalous behavior at T_c.

Substitution of Eqs. (43a) and (44) in Eq. (30b) gives the following expression for the time-correlation function:

$$S^{xx}(t) = \left(\frac{N}{2\beta\lambda'} - \frac{it\lambda}{2\hbar}(A\sigma - B_z^{-1})\right) \cos\left(\frac{\lambda\sigma}{\hbar}\right)t$$
$$+ \tfrac{1}{2}i\left(\tfrac{1}{2}N\sigma - A - \frac{t^2\lambda^2\sigma}{2B_z\hbar^2}\right) \sin\left(\frac{\lambda\sigma}{\hbar}\right)t. \quad (45a)$$

In the above expression, those containing t and t^2 come from the terms of N^{-1}. Hence, our results are valid for $0 \leq t < t_{max}$, where $t_{max} = (\hbar^2 N/\lambda^2)^{1/2}$. In this range of t, the correlation function is

$$S^{xx}(t) \approx \langle S_x^2 \rangle \cos(\lambda\sigma/\hbar)t + \tfrac{1}{2}i\langle S_z^2 \rangle^{1/2} \sin(\lambda\sigma/\hbar)t. \quad (45b)$$

In the low-temperature region ($T < T_c$) the correlation function is oscillatory, whereas in the high-temperature region the correlation function shows Gaussian behavior. Since $\sigma \to 0$ as $T \to T_c-$, it is evident that the Ising-like system exhibits critical slow down as in the high-temperature side. The form of critical slow down is, however, very different from the conventional form.[23] Finally, we note that in the isotropic limit (i.e., $\lambda \to 0$) both the XY- and Ising-correlation functions, in Eqs. (41b) and (45b) respectively, reduce to the same time-independent correlation function as is required.

V. CONCLUDING REMARKS

We have derived the transverse component of the time-correlation function $S^{xx}(t)$ for the spin Van der Waals system, valid to the time scale $t < O(N^{1/2})$ in an appropriate choice of units. Our results for the time-correlation function are not of exponential form as one might have expected from the similarity between the Van der Waals and Ornstein-Zernicke-type models.

As is already known, all the Van der Waals systems belong to one static universality class quite unlike the nearest-neighbor models. Our work shows that dynamic properties are, however, more subtle: For $T > T_c$ there is one dynamic class, but for $T < T_c$ there are two classes.

That there is but one static universality class for the Van der Waals system seems nevertheless reasonable. In the high-density limit, the spin interaction evidently is smoothed out, thereby masking spin dimensionality, at least to first order. But for dynamics, time evolution is still governed by the commutation relations of spin operators which are not affected by high densities. As a result, apparently not all subtle details of quantum origin are averaged out. For example, for the Ising-like system in the condensed phase, $S^{xx}(t)$ shows a kind of Larmor precession about the axis of the spontaneous magnetization $\langle S_z \rangle$. It precesses indefinitely and has a frequency which depends on the spontaneous magnetization. At high temperatures, the system ceases to precess as there is no longer any spontaneous magnetization. For the XY-like system in the condensed phase, the same correlation function cannot precess as in the Ising-like system, since the spontaneous magnetization now is also in the transverse direction. Thus, the time-correlation function smoothly approaches an equilibrium and it is also unaffected by the onset of critical fluctuations. Finally, since these dynamic features are consequences of the commutativity property of spin operators, it seems reasonable to conjecture that the nearest-neighbor models would have qualitatively similar dynamic behavior as described by the Van der Waals models.

[1]Y. de Ribaupierre and F. D. Manchester, J. Phys. C 7, 2126 (1974).

[2]C. Kittel and H. Shore, Phys. Rev. 138, A1165 (1965).

[3]Th. Niemeijer, Physica (Utr.) 48, 467 (1970).

[4]G. Vertogen and A. S. DeVries, Physica (Utr.) 59, 634 (1972).

[5]For these cooperative models with short-range interaction, it is now well established that static universality classes are determined by relevant variables, one of which is spin dimensionality. [See, for example, H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford U. P., Oxford, 1971)]. Thus, mean-field models evidently require much weaker static universality conditions.

[6]B. I. Halperin and P. C. Hohenberg, Phys. Rev. 177, 952 (1969).

[7]This work is also suggested by an earlier work on the dynamic behavior of the $S = \frac{1}{2}$ nearest-neighbor XY model. See M. H. Lee, Phys. Rev. B 8, 3290 (1973). It was shown there that under a high-frequency approximation the nearest-neighbor XY model becomes dynamically equivalent to the constant-coupling XY model.

[8]For the Ising-like case, our results given here may be regarded complementary to the longitudinal correlation function for $T \gtrsim T_c$ obtained by, e.g., K. Kawasaki and T. Yamada [Prog. Theor. Phys. 39, 1 (1968)], where the spin dynamics is generated by a master equation (i.e., a kinetic Ising model). The dynamic *critical* properties of such a stochastic system and other related ones are extensively reviewed recently by P. C. Hohenberg and B. I. Halperin [Rev. Mod. Phys. 49, 435 (1977)].

[9]A. Sur, D. Jasnow, and I. J. Lowe, Phys. Rev. B 12, 3845 (1975).

[10]U. Brandt and K. Jacoby, Z. Phys. B 25, 181 (1976).

[11]H. W. Capel and J. H. H. Perk, Physica (Utr.) 87A, 211 (1977).

[12]J. H. Van Vleck, *Electric and Magnetic Susceptibilities* (Clarendon, Oxford, 1932), pp. 323 and 324.

[13]R. H. Brout, *Phase Transitions* (Benjamin, New York, 1965), p. 11.

[14]That is, the entropy per spin $S_e/N = -k\,W(S) + k\,\ln 2 + O(N^{-1})$. Hence, $-k\,W(S)$ may be identified as our spin-dependent entropy term.

[15]A. Erdélyi, *Asymptotic Expansions* (Dover, New York, 1956), p. 36.

[16]Recall that S is obtained by vector addition of individual spins, i.e., $\vec{S} = \Sigma^N_{i=1} \vec{s}_i$ where $s_i = \frac{1}{2}$ for all i, and S_0 is one of the possible values of S. Evidently min S can result when individual spins are in a disordered state, whereas max S can result when individual spins are in an ordered state. Observe that $-W(S) \to 0$ as $S/N \to 0$ and $-W(S) < 0$ for $0 < S/N < \frac{1}{2}$ [see Eq. (8)].

[17]The specific heat vanishes to the order $O(N^{-1})$. For mean-field models the specific heat has a discontinuity at T_c.

[18]See M. H. Lee in Ref. 7.

[19]Near T_c, Eq. (16c) gives the familiar mean-field result $\sigma \sim (\beta/\beta_c - 1)^{1/2}$.

[20]M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* Natl.Bur. of Stds., Appl. Math. Ser. No. 55, (U. S. GPO, Washington, D.C., 1970), p. 298; *Higher Transcendental Functions*, Vol. II, edited by A. Erdélyi (McGraw-Hill, New York, 1953), p. 147.

[21]This result and also the similar one for the XY-like systems may be compared with the numerical work of D. Jasnow and M. Wortis [Phys. Rev. 176, 739, (1968)] for the nearest-neighbor anisotropic classical Heisenberg model. For the nearest-neighbor model, actually the exponent appears to behave universally but not the critical temperature.

[22]For mn models, one has similar equations of motion, which are examples of the generalized Langevin equation. These equations would be highly nonlinear and would not ordinarily be readily soluble except in some special limits. [See M. H. Lee and R. Dekeyser,

Physica (Utr.) 86–88 B+C, 1273 (1977), and references therein.] For Van der Waals models, however, the nonlinear or "random force" terms [the quadratic terms in Eq. (26)] are nonregenerative so that exact

general solutions of the time evolution can be obtained. [23]See, for example, W. Marshall and R. D. Lowde, Rep. Prog. Phys. 31, 705 (1968).