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Generalized Ornstein-Zernike equation

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A previously proposed generalization of the usual Ornstein-Zernike equation for near-critical correlations is examined. It is shown that near the critical point, to leading order, this equation is compatible with realistic critical behavior in both spatial and thermal variables. The scaling predictions of this equation are compared to known results for $d=2$ and $d=3$ Ising models. The generalized equation changes the usual Ornstein-Zernike values of various quantities in qualitatively the correct way; however, quantitative discrepancies remain.

I. INTRODUCTION

In a previous article[1] I have proposed a generalization of the traditional Ornstein-Zernike (OZ) differential equation for the pair correlation function of a fluid near its critical point. This generalization permits nonzero values of the critical exponent η and is therefore capable of describing correlations with realistic spatial behavior. I have shown in Ref. 1 that a certain long-distance asymptotic form for the triplet distribution function will, when placed in the second equation of the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy, yield this generalized Ornstein-Zernike (GOZ) equation. This paper presents some further results concerning the GOZ equation and the assumed asymptotic form for the triplet function.

In Sec. II, I consider the thermodynamic consistency of the asymptotic form near the critical point. The density derivative of the pair correlation is related to an integral of the triplet function through a sum rule. The leading contribution to this integral, near the critical point, can be evaluated from the asymptotic form of the triplet function. The asymptotic form is shown to satisfy the sum rule, to leading order, provided that the coefficient $a(s)$ of the leading term in the asymptotic form is given by $a(s)=\rho^{-1}(\partial f_2/\partial\rho)_\beta$, a result also obtained by others[2-4] who have investigated the very-long-range behavior of the triplet function in the critical region. When this $a(s)$ is used in the expression for κ (the inverse correlation length) derived in Ref. 1, there results a relation between κ and the isothermal compressibility K_T which is again consistent with realistic ($\eta\neq 0$) critical behavior. These results, along with those of Ref. 1, show that the proposed asymptotic form of the triplet function is consistent with realistic critical behavior in both spatial *and* thermal variables.

In Sec. III, I consider the scaling[5] form for the near-critical correlations predicted by the GOZ equation. The leading terms have the correct analytic form for both large and small values of the scaling variable $t=\kappa r$. The higher-order terms at small t do not have the expected[6] exponents. To get some idea of the numerical accuracy of the GOZ scaling function I have compared it to the known[7] result for the $d=2$ Ising model and to some numerical estimates[8] for the $d=3$ Ising models. In addition, I have considered the GOZ prediction for the ratios of the true to the second-moment inverse correlation lengths and compared these with known[9] results for Ising models. In all of these comparisons the GOZ results are shifted from the OZ results in the correct direction, but quantitative differences between the GOZ and Ising results remain. Finally, a brief discussion of the analytic behavior of the Fourier transform of the GOZ correlations is given. In Sec. IV these results are summarized and a brief discussion of possible improvement of the GOZ equation is given.

II. THERMODYNAMIC CONSISTENCY

Unless otherwise noted, the independent thermodynamic variables will be taken to be the number density ρ and the inverse temperature $\beta=1/kT$. The $f_n(\vec{r}_1,\ldots,\vec{r}_n)$ are the usual[1] reduced distribution functions. A standard[10] sum rule relating f_2 and f_3 is

$$\frac{\partial f_2(\vec{r}_1,\vec{r}_3)}{\partial\rho}=\frac{\beta}{\rho^2 K_T}\left\{2f_2(\vec{r}_1,\vec{r}_3)+\int[f_3(\vec{r}_1,\vec{r}_2,\vec{r}_3)-\rho f_2(\vec{r}_1,\vec{r}_3)]d\vec{r}_2\right\}, \qquad (2.1)$$

where $K_T^{-1}=\rho(\partial p/\partial\rho)_\beta$. As the critical point is approached, $K_T\to\infty$, but $\partial f_2/\partial\rho$ remains nonzero; hence the integral in (2.1) must diverge at the critical point. The divergent part of this integral can be found from the assumed[1] asymptotic expression for the triplet correlations. In terms of the dimensionless correlations

$$h_2(\vec{r}_1,\vec{r}_2)=[f_2(\vec{r}_1,\vec{r}_2)-\rho^2]/\rho^2$$

and

$$h_3(\vec{r}_1,\vec{r}_3\mid\vec{r}_2)=[f_2(\vec{r}_1,\vec{r}_2,\vec{r}_3)-\rho f_2(\vec{r}_1,\vec{r}_3)]/\rho^3 ,$$

the asymptotic form[1] is

$$h_3(\vec{r}_1,\vec{r}_3\mid\vec{r}_2)\simeq a(s)[1+b(s/r)^2+\cdots]h_2(r)/2+a(s)[1+b(s/t)^2+\cdots]h_2(t)/2 , \qquad (2.2)$$

where $\vec{r}=\vec{r}_1-\vec{r}_2$, $\vec{t}=\vec{r}_3-\vec{r}_2$, and $\vec{s}=\vec{r}_1-\vec{r}_3$. The form (2.2) is supposed to be valid for $s\sim r_0$ (the range of the intermolecular potential) and $r\gg r_0, t\gg r_0$, and has the required invariance under the interchange of $\vec{r}_1$ and $\vec{r}_3$. Higher-order terms in the small quantities $s/r, s/t$ have not been kept. An alternative form of (2.2) follows[1] from expanding $|\vec{t}|=|\vec{r}-\vec{s}|$ in powers of s/r and $h_2(t)$ in a Taylor's expansion about $t=r$,

$$h_3(\vec{r}_1,\vec{r}_3\,|\,\vec{r}_2)\simeq a(s)h_2(r)-\tfrac{1}{2}(\hat{r}\cdot\hat{s})a(s)\frac{dh_2(r)}{dr}+ba(s)(s/r)^2h_2(r)-\tfrac{1}{4}[1-(\hat{r}\cdot\hat{s})^2]a(s)(s/r)\frac{dh_2(r)}{dr}+\cdots . \tag{2.3}$$

In (2.3), I do not explicitly give higher-order terms in s/r and $dh_2(r)/dr$ since they do not affect the result. Now, in (2.1) I fix $s\equiv|\vec{r}_1-\vec{r}_3|\sim r_0$, change the variable of integration to $\vec{r}$, and split the integration into parts $r<R$ and $r>R$ where $R\gg r_0$ and is fixed. Thus (2.1) becomes

$$\frac{\partial f_2(s)}{\partial\rho}=\frac{\beta}{\rho^2K_T}\left[2f_2(s)+\rho^3\int_{r<R}h_3(\vec{r}_1,\vec{r}_3\,|\,\vec{r}_2)d\vec{r}+\rho^3\int_{r>R}h_3(\vec{r}_1,\vec{r}_3\,|\,\vec{r}_2)d\vec{r}\right] . \tag{2.4}$$

Only the last term in the large parentheses in (2.4) can diverge as the critical point is approached, and, in this term, the integrand can be approximated by (2.3) as follows:

$$\int_{r>R}h_3(\vec{r}_1,\vec{r}_3\,|\,\vec{r}_2)d\vec{r}=a(s)\int_{r>R}h_2(r)d\vec{r}-\tfrac{1}{2}a(s)\int(\hat{r}\cdot\hat{s})\frac{dh_2(r)}{dr}d\vec{r}+ba(s)\int_{r>R}(s/r)^2h_2(r)d\vec{r}-\tfrac{1}{4}a(s)\int_{r>R}[1-(\hat{r}\cdot\hat{s})^2](s/r)\frac{dh_2(r)}{dr}d\vec{r}+\cdots . \tag{2.5}$$

In (2.5), the last two integrals [and all other integrals corresponding to higher terms of (2.3)] are finite at the critical point if $\eta>0$, as is expected. Since the third integral in (2.5) vanishes because of the angular integration over $(\hat{r}\cdot\hat{s})$, the only divergent contribution comes from the second integral which, by the compressibility sum rule[11]

$$\rho K_T/\beta=1+\rho\int h_2(r)d\vec{r} , \tag{2.6}$$

diverges as $a(s)K_T/\beta$. These estimates for (2.5) allow (2.4) to be written as

$$\frac{\partial f_2(s)}{\partial\rho}=\frac{\beta}{\rho^2K_T}[a(s)\rho^3K_T/\beta+F(s)] , \tag{2.7}$$

where $F(s)$ is *finite* at the critical point but cannot be simply evaluated. Solving (2.7) for $a(s)$ gives

$$a(s)=\rho^{-1}\frac{\partial f_2(s)}{\partial\rho}-\beta F(s)/\rho^3K_T \tag{2.8}$$

near the critical point. This expresses the coefficient $a(s)$ of the asymptotic form (2.2) in terms of the pair distribution function and a contribution which vanishes as K_T^{-1} as the critical point is approached. This result, or its equivalent, can be found[2–4] in several discussions of the triplet distribution function.

Reference 1 provides an expression for the inverse correlation length κ in terms of integrals over $a(s)$, and thus it is interesting to see what Eq. (2.8) implies for κ. From Ref. 1, Eqs. (14), (11), and (10), we have[12]

$$\kappa^2=12(1-C_2R_0/2)/C_4R_2 , \tag{2.9}$$

$$C_2=\frac{2\pi^{d/2}}{d\Gamma(d/2)},\quad C_4=\frac{6\pi^{d/2}}{d(d+2)\Gamma(d/2)} , \tag{2.10}$$

$$R_n=\rho\int_0^\infty\frac{du(s)}{ds}a(s)s^{d+n}ds , \tag{2.11}$$

where $u(s)$ is the intermolecular potential. Now, κ^2 should vanish as the critical point is approached (as $K_T\to\infty$), and since R_2 in (2.9) is not divergent at the critical point, $1-C_2R_0/2$ should vanish as $K_T\to\infty$. This can be shown to follow from (2.8) by first writing κ^2, from (2.9)–(2.11), in the form

$$\kappa^2=\frac{12}{C_4R_2}\left[1-\frac{\rho}{2d}\int s\frac{du}{ds}a(s)d\vec{s}\right] \tag{2.12}$$

[the factor $C_2/2$ in (2.9) comes from the factor $1/2d$ and the angular integration in (2.12)]. The virial expression for the pressure[11] is

$$\beta p=\rho-\frac{1}{2d}\int s\frac{du}{ds}f_2(s)d\vec{s} . \tag{2.13}$$

This is differentiated with respect to ρ, and (2.8) is used to eliminate $\partial f_2(s)/\partial\rho$ to obtain

$$\beta\frac{\partial p}{\partial\rho}=1-\frac{1}{2d}\int s\frac{du}{ds}[\rho a(s)+\beta F(s)/\rho^2K_T]ds$$

or

$$1-\frac{\rho}{2d}\int s\frac{du}{ds}a(s)d\vec{s}=(\beta/\rho K_T)(1+F) , \tag{2.14}$$

where

$$F=\frac{1}{2d\rho}\int s\frac{du}{ds}F(s)d\vec{s} .$$

This, with Eq. (2.12), gives

$$\kappa^2=AK_T^{-1},\quad A=12\beta(1+F)/C_4R_2 , \tag{2.15}$$

which shows[13] that the κ^2 defined by (2.9) vanishes as $K_T \to \infty$. The factor A must also vanish at the critical point if (2.15) is to be consistent with expected critical behavior. To see this, note[14] that along the critical isochore, $K_T \sim \tau^{-\gamma}$, $\kappa \sim \tau^{\nu}$ [where $\tau = (T-T_c)/T_c$], and that one of the standard exponent relations is $\gamma = (2-\eta)\nu$ so that $A = K_T\kappa^2 \sim \tau^{\eta\nu} \sim \kappa^{\eta}$ and therefore vanishes as $\kappa \to 0$. (The result $K_T\kappa^2 \sim \kappa^{\eta}$ can also be derived from the compressibility sum rule by assuming the usual scaling form for the near-critical correlations.) The asymptotic form (2.2) does not determine the value of the factor F appearing in expression (2.15) for A; hence (2.2) does not imply that $A \sim \kappa^{\eta}$. On the other hand, Eq. (2.2) certainly appears consistent with the assumption that, at the critical point, $F \to -1$ in such a way that $A \sim \kappa^{\eta}$. Hence the conjectures of Ref. 1 concerning the asymptotic form of the near-critical triplet correlations appear consistent with realistic critical behavior in both spatial and thermal variables.

III. SCALING FUNCTION

In this section the GOZ scaling function[6,14] for the near-critical pair correlation is compared to various known results. The scaling function Γ is defined by assuming the correlations are of the form

$$h_2(r,\kappa) = \Gamma(\kappa r)/r^{d-2+\eta}, \quad \kappa r \ll 1, \; r \gg r_0 \tag{3.1}$$

where Γ is supposed to assume the asymptotic forms[6]

$$\Gamma(t) \simeq D - D_1 t^{(1-\alpha)/\nu} + \cdots \quad \text{as } t \to 0 \tag{3.2}$$

and

$$\Gamma(t) \simeq A(t^{d/2-3/2+\eta})e^{-t} \quad \text{as } t \to \infty . \tag{3.3}$$

The form (3.3) implies that κ in (3.1) is the true,[15] rather than the second-moment, inverse correlation length, and also implies that the correlation has the usual OZ form at large t. With this choice the scaling function Γ is believed to be universal[16] up to a multiplicative constant. Under this assumption a comparison of the GOZ prediction of Γ for fluid systems with known results for Ising models is appropriate.

The GOZ equation is[1,17]

$$\frac{d^2h(r)}{dr^2} + \frac{d-1}{r}\frac{dh(r)}{dr} - \frac{ph(r)}{r^2} - \kappa^2 h(r) = 0 \tag{3.4}$$

[the subscript of $h_2(r)$ will be suppressed], where the parameter p is assumed positive and determines the exponent η by

$$\eta(d-2+\eta) - p = 0 . \tag{3.5}$$

A straightforward substitution, $j = (\kappa r)^{d/2-1}h(r)$, shows that j satisfies the modified Bessels equation[18] of order

$$\mu = [(d-2)^2/4 + p]^{1/2} = d/2 - 1 + \eta \tag{3.6}$$

in the variable $t = \kappa r$. The solutions which vanish at $t = \infty$ are the McDonald functions[18] (modified Bessel function of the second kind), and hence the GOZ equation has solutions

$$h(r) \alpha t^{1-d/2} K_\mu(t) . \tag{3.7}$$

The asymptotic[18] forms for $K_\mu(t)$ are

$$K_\mu(t) \simeq \frac{\pi}{2\sin(\mu\pi)}(t/2)^{-\mu}\left[\frac{1}{\Gamma(1-\mu)} + \frac{(t/2)^2}{\Gamma(2-\mu)} + \cdots\right] \quad t \to 0, \; \mu \neq \text{integer} \tag{3.8}$$

and

$$K_\mu(t) \sim (\pi/2t)^{1/2} e^{-t} \quad \text{as } t \to \infty . \tag{3.9}$$

Comparison of (3.7) with (3.1) gives the GOZ scaling function

$$\Gamma_G = C t^{\mu} K_\mu(t), \quad \mu = d/2 - 1 + \eta \tag{3.10}$$

where C is an undetermined constant. From (3.8)—(3.10) the GOZ values for the constants D and A in (3.2) and (3.3) are

$$D_G = C\pi 2^{\mu-1}/\Gamma(1-\mu)\sin(\mu\pi), \quad A_G = C(\pi/2)^{1/2} , \tag{3.11}$$

giving a (supposedly universal) ratio of

$$D_G/A_G = \pi^{1/2} 2^{\mu-1/2}/\Gamma(1-\mu)\sin(\mu\pi) . \tag{3.12}$$

Equations (3.7) and (3.8) also show that the GOZ equation predicts the exponent of the correction term in (3.2) to be 2 and independent of dimension. The correct exponent[6] is believed to be $(1-\alpha)/\nu$, which has the values 1 in $d=2$ and ~ 1.4 in $d=3$ Ising models. Hence the correction exponent in (3.2) predicted by the GOZ equation is too large.

The GOZ prediction (3.12) for the amplitude ratio and, in some cases, (3.10) for the scaling function, can be compared to known Ising-model results. For the two-dimensional Ising model the amplitude ratio is known[19] to be

$$D/A = 0.7034(\pi^{1/2})(2^{1/8}) = 1.36, \quad d=2 \tag{3.13}$$

when the critical point is approached in zero field from high temperatures. The GOZ prediction (3.12), with the known values $d=2$, $\eta=\frac{1}{4}$, and $\mu=\frac{1}{4}$, is

$$D_G/A_G = 1.72 , \tag{3.14}$$

and hence the GOZ amplitude ratio is about 26% too large for $d=2$. This effect is numerically much larger than the previously noted error in the correction terms at small t. A comparison of the numerical values of the actual[20] $d=2$ Ising scaling function with the GOZ values is shown in Table I. The value of C in (3.10) has been chosen so that the two scaling functions match at $t=8$.

Similar, though numerically smaller, discrepancies are found for the $d=3$ Ising model where the D and A coefficients are known from high-temperature-series extrapolations for several lattices.[8] That the same value,

$$D/A = 0.873 \pm 0.01, \quad d=3 \tag{3.15}$$

is found for the fcc, bcc, and sc lattices is an indication of the expected universality of this ratio. The GOZ prediction (3.12) depends on the value of μ and therefore η. The precise value of η is perhaps still uncertain,[21] but for this comparison I will use $\eta = 0.04$, which gives

TABLE I. Comparison of the $d=2$ Ising scaling function (zero field, $T>T_c$) with the GOZ scaling function. (Diff. denotes difference.)

t	$\Gamma(t)$	$\Gamma_G(t)$	Diff. (%)
8	1.0169×10^{-4}	1.0169×10^{-4}	0
5	2.278×10^{-3}	2.283×10^{-3}	0.2
3	1.887×10^{-2}	1.898×10^{-2}	0.6
1	1.738×10^{-1}	1.772×10^{-1}	2
0.1	0.5699	0.6210	9
0.05	0.6244	0.6918	12
0.01	0.6819	0.8019	18
0.002	0.6980	0.8487	22

$$D_G/A_G=0.954,\quad d=3,\ \eta=0.04\ . \tag{3.16}$$

Again, the GOZ value is too high (by about 9%), but is an improvement over the usual OZ value of 1 for this ratio.

The GOZ equation also allows a computation of the ratio of the true to the second-moment inverse correlation range, κ/κ_1. It is κ that appears as a parameter in the GOZ equation [Eq. (3.4)], while κ_1 is defined by the second moment of the correlation function as

$$\kappa_1^{-2}=\frac{\frac{1}{2}\int h(r)r^2\cos^2\theta\, d\vec{r}}{\int h(r)d\vec{r}}\ . \tag{3.17}$$

The angular integrals in (3.17) give a factor of $1/d$, and when the GOZ scaling form [Eqs. (3.1) and (3.10)] is used for $h(r)$, (3.17) becomes

$$(\kappa^2/\kappa_1^2)_G=\frac{\int_0^\infty K_\mu(t)t^{d/2+2}dt}{2d\int_0^\infty K_\mu(t)t^{d/2}dt}\ . \tag{3.18}$$

This ratio of integrals can be evaluated by multiplying the modified Bessel equation for $K_\mu(t)$ by $t^{d/2+2}$ and integrating. A straightforward calculation yields

$$(\kappa^2/\kappa_1^2)_G=\frac{(d/2+1)^2-\mu^2}{2d}=1-\frac{\eta(d-2)+\eta^2}{2d}\ . \tag{3.19}$$

For the usual OZ theory ($\eta=0$) this ratio is 1. The GOZ value of this ratio is 0.984 for $d=2$ and $\eta=\frac{1}{4}$, and 0.993 for $d=3$ and $\eta=0.04$. The Ising values[9] appear to depend somewhat on the method used to extract them. For both $d=2$ and $d=3$ lattices, however, the ratio κ^2/κ_1^2 is probably greater than 0.999 and less than 1. The GOZ prediction is qualitatively correct, even though the change from the OZ value of 1 is perhaps too large.

Critical correlations are often[6,15,9,22] discussed in terms of their Fourier transform, defined by

$$\chi(\vec{k},\kappa)=\int e^{i\vec{k}\cdot\vec{r}}h_2(r,\kappa)d\vec{r}\ .$$

The scaling form,[22] in the variable $y=\vec{k}/\kappa$, is $\chi(k,\kappa)=\kappa^{\eta-2}X(y)$, where, from (3.7),

$$X(y)=\int e^{i\vec{y}\cdot\vec{t}}t^{1-d/2}K_\mu(t)d\vec{t}\ . \tag{3.20}$$

The angular integrals in (3.20) can be done, reducing the integral to a Fourier sine transform which can be evaluated[23] in terms of a hypergeometric function of argument $-y^2$. From the known[23] analytic properties of the hypergeometric function one can show the Fourier transform (3.20) of the GOZ scaling function has singularities only at $y=\pm i$ and at infinity with a branch cut conventionally chosen on the imaginary axis. The details of the singularity structure at $y=\pm i$ depend only on the dimension d, while the order of the branch points at infinity depend on both d and the exponent η. The singularity structure of the GOZ approximation is certainly simpler than that of the exact $d=2$ Ising model[22] which has an infinite sequence of branch points on the imaginary axis, but is richer than that of the usual OZ theory which has only simple poles at $y=\pm i$. It is perhaps closer to, although not identical with, the Fisher-Burford[9] approximate in its analytic structure. The Fisher-Burford approximate, however, has one more free parameter than the GOZ, which allows independent amplitudes at both large and small y.

IV. CONCLUSIONS

As a phenomenological description of near-critical correlations, this generalization of the Ornstein-Zernike theory is, in several ways, more satisfactory than the usual Ornstein-Zernike theory. There remain, however, serious deficiencies. On the positive side, the GOZ has the correct analytic behavior, to leading order, at both large and small values of the spatial scaling variable κr and appears to be at least consistent with realistic critical behavior in the thermal variables as well. The amplitude ratios and the scaling function are shifted from the usual OZ theory in the correct direction for $\eta>0$. On the negative side, the corrections to the leading order of the scaling function at small κr have the wrong exponents and the amplitude ratios are not quantitatively correct. Finally, for Ising systems, there are lines of states in the thermodynamic plane ($H=0$, $T<T_c$) along which the pair correlations do not have[24] the OZ form at large κr, and, presumably, such lines exist for fluids as well. For these states the amplitude of the leading OZ term at large κr has vanished and correction terms dominate the large-κr behavior. These terms are not contained in the GOZ.

What are the prospects for improving the GOZ? It seems clear from the "derivation" of Ref. 1 that the GOZ results from an asymptotic expansion in the two small parameters κr_0 and r/r_0 and that higher-order terms in these parameters will produce corrections to the GOZ theory. It seems unlikely to me that these effects will be capable of correcting the above-noted deficiencies of the theory. What appears more likely[25] is that a small but non-negligible coupling between density and energy-density fluctuations must be taken into account even in the lowest-order theory. In such a theory, one might expect, instead of the GOZ, a coupled set of three differential equations for the density-density, the density—energy-density and the energy-density—energy-density correlations. The form that such equations should take and their resulting scaling properties is not known.

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