Journal of Statistical Physics, Vol. 72, Nos. 5/6, 1993

Hierarchical Reference Theory of Fluids: Application to Three-Dimensional Ising Model

D. Pini,[1] A. Parola,[1] and L. Reatto[1]

Received January 11, 1993

The hierarchical reference theory (HRT) of fluids is applied to the three-dimensional Ising model on a simple cubic lattice with nearest-neighbor ferromagnetic interaction via the equivalence with the lattice-gas model. The hierarchy is truncated to the first equation and closed with an Ornstein–Zernike ansatz for the direct correlation function embodying both thermodynamic consistency and on-site repulsion between lattice particles. The resulting equations are integrated numerically above and below the critical temperature and the results are compared with those obtained by closed-form approximants. We show that HRT yields nontrivial critical exponents with the correct scaling regime and a value of the critical temperature in very close agreement with the true one. At the same time it retains all the information about the short-range behavior of the system, and so gives a very accurate description also away from the critical point. Below the critical temperature as long as long-wavelength fluctuations are included in the system the van der Waals loop is suppressed and is replaced by a region where the compressibility is infinite, namely the coexistence region.

KEY WORDS: Ising model; direct correlation function; critical behavior; coexistence region.

1. INTRODUCTION

A realistic description of the critical behavior of a fluid requires a careful treatment of the long-range fluctuations arising when we approach the critical point. At the same time the short-range correlations, which depend on the specific form of the interparticle interaction, affect nonuniversal properties of the system and dominate away from the critical region. It is then clear that if we want to achieve a good description of a fluid over the whole phase plane, we must develop a theory able to deal with charac-

---

[1] Dipartimento di Fisica, Università degli Studi di Milano, 20133 Milan, Italy.

0022-4715/93/0900-1179$07.00/0

teristic lengths varying over a very wide range. The hierarchical reference theory (HRT)^(1,2) is a microscopic theory of fluids explicitly devised to pursue such a nontrivial task. In this theory the attractive part of the interaction is turned on gradually in such a way that its long-wavelength Fourier components responsible for the critical behavior are included in the system only in the final stage of the process. The corresponding evolution of the free energy and of the correlation functions of increasing order is then expressed by an infinite hierarchy of exact integrodifferential equations.^(1) The HRT shares with the renormalization group the idea of approaching the real system through a process in which fluctuations of increasing wavelength are gradually taken into account. On the other hand, it is based on the microscopic Hamiltonian of the system and retains all the information about short-range properties important away from the critical point and can thus be regarded as a genuine microscopic theory of the fluid state.

It was previously shown^(1) that a simple closure of the hierarchy assuming that the direct correlation function of the system is analytic in the wavevector k even at the critical point (the Ornstein–Zernike ansatz) yields nonclassical critical exponents with the correct scaling regime. Later^(2) the same kind of closure was extended away from the critical region and the theory was applied to simple fluids above the critical temperature. The results were found to be in good agreement both with numerical simulations and experiments.

It was already pointed out^(1) that this theory could be reformulated for a system of particles on a lattice and could thus be applied to an Ising spin system by exploiting the well-known equivalence (see, e.g., ref. 3) between this model and the lattice gas. In the present work the HRT with a closure of the same class as the previous ones has been applied to the three-dimensional ferromagnetic Ising model on a simple cubic lattice with nearest-neighbor interaction. This application is of some interest because in the case of real systems comparing theoretical predictions with experimental data may be not so straightforward due to the difficulty of modeling such systems with two-body interactions. The comparison with numerical simulations overcomes this difficulty, but finite-size effects make them not suited for use inside the critical region. For these reasons it is not very easy, e.g., to establish how accurately the critical temperature is predicted by the theory. For the Ising model, on the other hand, these difficulties do not arise because the predictions of the theory can be compared both inside and outside the critical region with a great deal of highly reliable results, such those yielded by approximants based on extrapolations of series expansions.^(4,5)

Moreover, in the present work our theory has been applied also below the critical temperature. This point is rather relevant because the first-order phase transition taking place in this region is considerably less investigated than the second-order one at the critical point. Most of the theories are not able to reproduce the strongly nonanalytic behavior of the thermodynamic functions below T_c and this gives rise inside the coexistence region to a negative value of the compressibility and hence to the so-called van der Waals loop, which is then artificially eliminated via the *ad hoc* Maxwell construction. In this work we will show that the HRT not only describes the system very accurately above T_c, but is indeed able to suppress such unphysical behavior and to yield an infinite value for the compressibility inside the whole coexistence region.

The paper is organized as follows: in Section 2 the theory is reviewed and then the application to the lattice gas is described, in Sections 3 and 4 the results obtained respectively above and below the critical temperature, expressed in terms of the usual magnetic quantities, are discussed and compared both with other theories and with closed-form approximants, and finally in Section 5 our conclusions are reported. In the Appendix our equations are derived in detail, paying particular care to the exact implementation of the core condition.

2. THEORY

We consider a homogeneous system of particles interacting by a two-body potential

$$v(\mathbf{r}) = v_R(\mathbf{r}) + w(\mathbf{r}) \tag{1}$$

where $v_R(\mathbf{r})$ contains the singular part of the interaction due to the short-range repulsion between particles, while $w(\mathbf{r})$ is the attractive part which dominates at large distance. We assume that the properties of the system interacting via the potential $v_R(\mathbf{r})$ alone are known, so we can regard it as the "reference" system. Our aim is to treat accurately the long-range fluctuations responsible for the critical behavior of the completely interacting system: in the HRT this is achieved by turning on the attractive part of the interaction gradually, starting from shorter length scales. To this purpose, let us introduce a region Ω_Q labeled by a parameter Q which, during the evolution of Q, spans the whole momentum space from large to small wavevectors $\mathbf{k}$, shrinking on the origin $\mathbf{k} = \mathbf{0}$ at the end of the process. We can now define a potential $w_Q(\mathbf{r})$ via its Fourier transform $\tilde{w}_Q(\mathbf{k})$ in the following way:

$$\tilde{w}_Q(\mathbf{k}) = \begin{cases} 0, & \mathbf{k} \in \Omega_Q \\ \tilde{w}(\mathbf{k}), & \mathbf{k} \notin \Omega_Q \end{cases} \tag{2}$$

where $\tilde{w}(\mathbf{k})$ is the Fourier transform of the full attractive potential $w(\mathbf{r})$. Then Q plays the role of a long-wavelength cutoff; for example, the domain Ω_Q could correspond to $k < Q$ and actually this would be the most natural choice in the case, different from the present one, of a system of particles interacting by a spherically symmetric potential, like the simple fluids studied in ref. 2. As Q evolves from its initial value Q_i to its final value Q_f, the fully interacting system is approached, starting from the reference one, through a sequence of Q-systems with potentials given by

$$v_Q(\mathbf{r}) = v_R(\mathbf{r}) + w_Q(\mathbf{r}) \tag{3}$$

The effect of the interaction between two Q-systems infinitesimally close to each other can be determined exactly by perturbation theory. This enables us to study the evolution of the free energy and of the correlation functions of the system as the interaction takes on its long-wavelength Fourier components. This evolution is described by an exact hierarchy of integro-differential equations involving correlation functions of increasing order. The first two equations of the hierarchy read(1)

$$\frac{\partial \mathscr{A}_Q}{\partial Q} = -\frac{1}{2}\int_{\Sigma_Q} \frac{d\omega_k}{(2\pi)^3} \log\left(1 - \frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})}\right) \tag{4}$$

$$\begin{aligned}\frac{\partial \mathscr{C}_Q}{\partial Q}(\mathbf{p}) = -\int_{\Sigma_Q} d\omega_k \Bigg\{ \Bigg[&\frac{1}{2} c_4^Q(\mathbf{p}, -\mathbf{p}, \mathbf{k}, -\mathbf{k}) \\ &- \frac{c_3^Q(\mathbf{p}, \mathbf{k}, -\mathbf{p}-\mathbf{k})\, c_3^Q(-\mathbf{p}, -\mathbf{k}, \mathbf{p}+\mathbf{k})}{c_Q(\mathbf{p}+\mathbf{k})} \Bigg] \\ &\times \frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})[\mathscr{C}_Q(\mathbf{k}) - \tilde{\Phi}(\mathbf{k})]} \Bigg\}\end{aligned} \tag{5}$$

In these equations $c_Q(\mathbf{k})$ and $c_n^Q(\mathbf{k}_1,..., \mathbf{k}_n)$ are respectively the two- and n-particle direct correlation functions of the Q-system in momentum space, including the ideal gas contribution. $c_Q(\mathbf{k})$ is related to the usual direct correlation function $c_Q^{\mathrm{OZ}}(\mathbf{k})$ defined by the Ornstein–Zernike relation by

$$c_Q^{\mathrm{OZ}}(\mathbf{k}) = \frac{1}{\rho} + c_Q(\mathbf{k}) \tag{6}$$

where ρ is the density of the system. The integrals on the right-hand sides of (4), (5) are evaluated on the boundary Σ_Q of the region Ω_Q, $d\omega$ is the surface element spanning Σ_Q, and $\tilde{\Phi}(\mathbf{k})$ is the Fourier transform of the quantity $\Phi(\mathbf{r})$ defined by

$$\Phi(\mathbf{r}) = -\frac{w(\mathbf{r})}{kT} \tag{7}$$

The quantities $\mathscr{A}_Q$ and $\mathscr{C}_Q(\mathbf{k})$ are related, respectively, to the free energy per unit volume a_Q and to the direct correlation function in momentum space $c_Q(\mathbf{k})$ of the Q-system, the difference being that in the "modified" quantities the discontinuous contribution due to $\tilde{\Phi}_Q(\mathbf{k})$ has been eliminated:

$$\mathscr{A}_Q = -\frac{a_Q}{kT} - \frac{1}{2}\rho[\Phi(\mathbf{0}) - \Phi_Q(\mathbf{0})] + \frac{1}{2}\rho^2[\tilde{\Phi}(0) - \tilde{\Phi}_Q(0)] \tag{8}$$

$$\mathscr{C}_Q(\mathbf{k}) = c_Q(\mathbf{k}) + \tilde{\Phi}(\mathbf{k}) - \tilde{\Phi}_Q(\mathbf{k}) \tag{9}$$

We recall that the direct correlation function in momentum space $c_Q(\mathbf{k})$ is related to the usual structure factor $S_Q(\mathbf{k})$ by the relation $c_Q(\mathbf{k}) = -1/\rho S_Q(\mathbf{k})$. When Q reaches its final value Q_f we have $\Phi_{Q_f}(\mathbf{r}) = \Phi(\mathbf{r})$, so that $-kT\mathscr{A}_{Q_f}$ and $\mathscr{C}_{Q_f}(\mathbf{k})$ coincide, respectively, with the free energy per unit volume a and with the direct correlation function $c(\mathbf{k})$ of the fully interacting system. At the beginning of the evolution of Q, on the other hand, $\Omega_Q = \Omega_{Q_i}$ covers the entire momentum space and hence we have $\Phi_{Q_i}(\mathbf{r}) = 0$, $a_{Q_i} = a_R$, $c_{Q_i}(\mathbf{k}) = c_R(\mathbf{k})$, where a_R and $c_R(\mathbf{k})$ are the free energy per unit volume and the direct correlation function of the reference system. The modified quantities $\mathscr{A}_{Q_i}$ and $\mathscr{C}_{Q_i}(\mathbf{k})$ then yield the usual random phase approximation to a and $c(\mathbf{k})$.

We want to apply our theory to the three-dimensional Ising model on a simple cubic lattice with ferromagnetic, nearest-neighbor interaction. The Hamiltonian is

$$\mathscr{H}_{\mathrm{Is}} = -J \sum_{\langle i,j \rangle} s_i s_j - H \sum_j s_j \tag{10}$$

where the first sum is over nearest-neighbor sites, $J > 0$ is the coupling constant, $s_i = \pm 1$ is the spin variable on the i site, and H is the external magnetic field. We will exploit the standard result (e.g., ref. 3) that this model can be mapped into a fluid of particles on a lattice interacting by the potential $v(\mathbf{r}_i - \mathbf{r}_j)$ defined as

$$v(\mathbf{r}_i - \mathbf{r}_j) = \begin{cases} +\infty & \mathbf{r}_i = \mathbf{r}_j \\ -w < 0 & i, j \text{ nearest neighbors} \\ 0 & \text{otherwise} \end{cases} \tag{11}$$

so that to every "magnetic" quantity related to the Ising model we can associate a corresponding "fluid" quantity. For example, J, H, and the magnetization per site M are given by

$$J = \frac{w}{4} \tag{12}$$

$$H = -\frac{1}{2}\left(\mu + \frac{qw}{2}\right) \tag{13}$$

$$M = 1 - 2\rho \tag{14}$$

where q is the coordination number and μ is the chemical potential of the lattice fluid. Here we assume the lattice spacing to be unity, so that the density ρ is the average number of particles per site. In the following we will refer to the "fluid" picture of the system for the application of the theory, and will then restate our results in magnetic terms.

In (11) it is immediate to identity the reference potential $v_R(\mathbf{r})$ with the singular on-site repulsion causing each site to be occupied by at most one particle, and the perturbation with the attractive nearest-neighbor contribution. We have then

$$\tilde{\Phi}(\mathbf{k}) = \lambda\gamma(\mathbf{k}) \tag{15}$$

where we have defined

$$\gamma(\mathbf{k}) = \frac{1}{3}(\cos k_x + \cos k_y + \cos k_z) \tag{16}$$

$$\lambda = \frac{6w}{kT} \tag{17}$$

and $\mathbf{k}$ varies in the first Brillouin zone. The reference system is nothing but a hard-sphere lattice gas, which corresponds to a system of noninteracting spins. This system is immediately solvable and one obtains

$$\frac{a_R}{kT} = \rho \log \rho + (1-\rho)\log(1-\rho) \tag{18}$$

$$c_R(\mathbf{k}) = -\frac{1}{\rho(1-\rho)} \tag{19}$$

In this work, as in the previous ones,$^{(1,2)}$ the hierarchy has been truncated to the first equation [Eq. (4)], relating the evolution of the free energy of the Q-system to the corresponding two-particle direct correlation function. In order to get a closed equation for the free energy we must supplement Eq. (4) with a closure relation involving the correlation function $\mathscr{C}_Q(\mathbf{k})$. An approximation which is widely used in the theory of fluids is the optimized random-phase approximation (ORPA).[2] In terms of the direct

---

[2] See, e.g., ref. 6, and refs. 19 and 21 for the application to the lattice gas.

correlation function $c(\mathbf{k})$ of the fully interacting system this amounts to setting

$$c(\mathbf{k}) = c_R(\mathbf{k}) + \tilde{\Phi}(\mathbf{k}) + \mathscr{G}(\mathbf{k}) \tag{20}$$

where the quantity $\mathscr{G}(\mathbf{k})$ is determined for given density and temperature in such a way that the two-particle radial distribution function $g(\mathbf{r})$ vanishes for small enough $\mathbf{r}$ in order to take into account the effect of the singular repulsive interaction $v_R(\mathbf{r})$. In the case of the lattice system with which we are dealing, this constraint, known as the *core condition*, involves only the point $\mathbf{r} = \mathbf{0}$ and this implies that $\mathscr{G}$ is independent of $\mathbf{k}$. For our closure relation we have chosen a sort of ORPA-like expression$^{(2)}$

$$\mathscr{C}_Q(\mathbf{k}) = c_R(\mathbf{k}) + \tau_Q \tilde{\Phi}(\mathbf{k}) + \sigma_Q \tag{21}$$

where τ_Q and σ_Q are unknown functions of density and temperature for every Q-system. Since in our case the direct correlation function of the reference system $c_R(\mathbf{k})$ does not actually depend on $\mathbf{k}$ [see Eq. (19)], we can write

$$\mathscr{C}_Q(\mathbf{k}) = E_Q(\rho, T) + B_Q(\rho, T)[1 - \gamma(\mathbf{k})] \tag{22}$$

In order to determine the unknown quantities $E_Q(\rho, T)$ and $B_Q(\rho, T)$, we require that our closure relation (22) satisfies two conditions which are related respectively to the long- and to the short-range behavior of the system, namely the thermodynamic consistency condition and the core condition we have just mentioned.

For the theory to be thermodynamically consistent, the structure factor $S_Q(\mathbf{k})$ must satisfy the well-known compressibility sum rule for every Q-system. In terms of $\mathscr{C}_Q(\mathbf{k})$ this condition reads

$$\mathscr{C}_Q(\mathbf{k} = 0) = E_Q(\rho, T) = \frac{\partial^2 \mathscr{A}_Q}{\partial \rho^2} \tag{23}$$

If we use this condition in Eq. (4) we get a partial differential equation for the evolution of the free energy involving $\partial \mathscr{A}_Q / \partial Q$ and $\partial^2 \mathscr{A}_Q / \partial \rho^2$. We expect the critical behavior yielded by the theory to be governed by the small-wavevector limit of $\mathscr{C}_Q(\mathbf{k})$ via the right-hand side of (4). In this limit Eq. (22) becomes

$$\mathscr{C}_Q(\mathbf{k}) \sim \frac{\partial^2 \mathscr{A}_Q}{\partial \rho^2} + \frac{B_Q}{6} k^2 \tag{24}$$

At the critical point the inverse compressibility is zero and (24) yields $c(\mathbf{k}) \sim k^2$, provided B_Q tends to a finite limit. The closure relation (22) implies then $\eta = 0$, i.e., the Ornstein–Zernike approximation: this can be somewhat justified by the small value of η in three dimensions. In ref. 1 it was shown that the thermodynamic consistency requirement and the Ornstein–Zernike approximation fixing the small-k behavior of $\mathscr{C}_Q(\mathbf{k})$ unambiguously determine the universal properties at the critical point predicted by the theory. An asymptotic analysis near the critical point in the spirit of the renormalization group shows that the HRT with an Ornstein–Zernike closure gives the correct critical exponents to linear order in ε ($\varepsilon = 4 - d$) in an expansion in dimensionality d, and in $d = 3$ the exponents have the values

$$\nu = 0.689, \qquad \gamma = 1.378, \qquad \beta = 0.345, \qquad \delta = 5 \tag{25}$$

They differ from the correct values by about 10% and satisfy the scaling laws with $\eta = 0$. We have explicitly checked (see Section 3) that the critical exponents obtained in the present case agree with those in (25) given by the "asymptotic" theory.

We now come to the core condition: in our case this requires that the two-particle radial distribution function $g_Q(\mathbf{r})$ vanishes at $\mathbf{r} = \mathbf{0}$ for every Q-system due to the singular on-site repulsion $v_R(\mathbf{r})$ contained in the interaction $v_Q(\mathbf{r})$. This condition can be written in terms of the direct correlation function $c_Q(\mathbf{k})$ of the Q-system and hence, using (9), in terms of the modified quantity $\mathscr{C}_Q(\mathbf{k})$ to give

$$\int_{\Omega_Q} \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{1}{\mathscr{C}_Q(\mathbf{k}) - \tilde{\Phi}(\mathbf{k})} + \int_{\mathscr{B} - \Omega_Q} \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{1}{\mathscr{C}_Q(\mathbf{k})} = \rho^2 - \rho \tag{26}$$

where $\mathscr{B}$ is the first Brillouin zone of the lattice.

Equation (4) and the closure relation (22) with the conditions (23), (26) give a closed system of two nonlinear integrodifferential equations; our aim is now to cast this system in a more tractable form. We observe that due to the form (22) for $\mathscr{C}_Q(\mathbf{k})$, the integrand on the right-hand side of Eq. (4) depends on $\mathbf{k}$ only via the nearest-neighbor Fourier transform $\gamma(\mathbf{k})$, so that it is natural to choose the integration surfaces Σ_Q as those defined by $\gamma(\mathbf{k}) = Q$, $-1 \leqslant Q \leqslant 1$. The most straightforward way to introduce the fluctuations in the system would then be to start from the boundary of the Brillouin zone and to go inside. If we do this, however, the evolution of the free energy described by Eq. (4) turns out to be unstable over the whole region where $\gamma(\mathbf{k})$ is negative, which corresponds to the first half of the integration path. This is a purely technical problem, which nevertheless does not allow for a direct numerical implementation of this procedure. In order to obtain the stability needed for numerical integration, the attractive part of the interaction has been introduced in the system starting from the surface $\gamma(\mathbf{k})=0$ inside the Brillouin zone and then moving toward the center and the boundary of the zone at the same time. This is achieved by choosing the region Ω_Q in Eq. (2) as given by $|\gamma(\mathbf{k})|>-Q$, $-1\leqslant Q\leqslant 0$. With this choice Eq. (4) becomes

$$\frac{\partial \mathscr{A}_Q}{\partial Q}=-\frac{1}{2}F(Q)\left[\log\left(1-\frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})}\right)\bigg|_{\gamma(\mathbf{k})=Q}+\log\left(1-\frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})}\right)\bigg|_{\gamma(\mathbf{k})=-Q}\right] \tag{27}$$

where $F(Q)$ is the density of states (see, e.g., ref. 7)

$$F(Q)=\int_{\mathscr{B}}\frac{d^3\mathbf{k}}{(2\pi)^3}\,\delta(Q-\gamma(\mathbf{k})) \tag{28}$$

Since the core condition (26) must be satisfied for every Q, we can differentiate it with respect to Q to get

$$\begin{aligned}\int_{\Omega_Q}&\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{[\mathscr{C}_Q(\mathbf{k})-\tilde{\Phi}(\mathbf{k})]^2}\frac{d\mathscr{C}_Q}{dQ}(\mathbf{k})+\int_{\mathscr{B}-\Omega_Q}\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{\mathscr{C}_Q^2(\mathbf{k})}\frac{d\mathscr{C}_Q}{dQ}(\mathbf{k})\\ &=F(Q)\left[\frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})[\mathscr{C}_Q(\mathbf{k})-\tilde{\Phi}(\mathbf{k})]}\bigg|_{\gamma(\mathbf{k})=Q}\right.\\ &\qquad\left.+\frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})[\mathscr{C}_Q(\mathbf{k})-\tilde{\Phi}(\mathbf{k})]}\bigg|_{\gamma(\mathbf{k})=-Q}\right]\end{aligned} \tag{29}$$

The boundary term on the right-hand side of Eq. (29) is due to the discontinuity of the "true" direct correlation function $c_Q(\mathbf{k})$ on the surface Σ_Q [see Eq. (9)].

In the previous work on simple fluids$^{(2)}$ the core condition was treated with an approximate scheme by decoupling small and large wavevectors $\mathbf{k}$. In our case, on the other hand, the core condition can be handled more easily than in a real fluid because it involves, for every Q-system, only one point in the coordinate space instead of a finite interval and therefore can be implemented exactly (of course within the limits in accuracy implied by the numerical calculation). Our approach relies on the fact that Eq. (27) can be cast in the following form:

$$\frac{\partial}{\partial Q}\left(\frac{\partial^2\mathscr{A}_Q}{\partial\rho^2}\right)=-\frac{1}{2}Q^2F(Q)\frac{\partial^2 v_Q}{\partial\rho^2} \tag{30}$$

which is obtained from Eq. (27) by differentiating twice with respect to ρ and defining $v_Q(\rho, T)$ as

$$v_Q=\frac{1}{Q^2}\left[\log\left(1-\frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})}\right)\bigg|_{\gamma(\mathbf{k})=Q}+\log\left(1-\frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})}\right)\bigg|_{\gamma(\mathbf{k})=-Q}\right] \tag{31}$$

If we combine Eqs. (29)–(31) and use the closure relation (22) for $\mathscr{C}_Q(\mathbf{k})$ we get a system of two differential equations in quasilinear form:

$$\frac{\partial^2 v_Q}{\partial \rho^2} = C(Q, v_Q, B_Q)\frac{\partial v_Q}{\partial Q} + D(Q, v_Q, B_Q) \tag{32}$$

$$\frac{\partial B_Q}{\partial Q} = K(Q, v_Q, B_Q)\frac{\partial v_Q}{\partial Q} + L(Q, v_Q, B_Q) \tag{33}$$

where B_Q has been defined in Eq. (22). The detailed calculation and the explicit form of the coefficients C, D, K, and L are reported in the Appendix. Once the quantities v_Q and B_Q have been obtained, we can get $\partial^2 \mathscr{A}_Q/\partial \rho^2$ and hence the compressibility of the system by inverting Eq. (31). The integration of Eqs. (32), (33) must be performed from $Q = 0$ to $Q = -1$, which correspond respectively to the reference and to the fully interacting system. From the exact expression (19) for the direct correlation function of the reference system, we get the following set of initial conditions:

$$v_{Q=0} = \lambda^2[\rho(1-\rho)]^2 \tag{34}$$

$$B_{Q=0} = -\lambda \tag{35}$$

with λ given by Eq. (17). The partial differential equation (32) must also be supplemented with suitable boundary conditions for the low- and high-density behavior of the quantity v_Q. At zero density the direct correlation function $c_R(\mathbf{k})$ of the reference system diverges [see Eq. (19)], so that Eqs. (9) and (31) yield

$$v_Q(\rho = 0) = 0 \qquad \text{for every } Q \tag{36}$$

Moreover, Eqs. (32), (33) preserve the symmetry with respect to $\rho = 1/2$ shown in the initial conditions (34), (35), so that the integration with respect to the variable ρ can be restricted to the interval (0, 1/2) with the condition

$$v_Q(\rho) = v_Q(1-\rho) \qquad \text{for every } Q \tag{37}$$

The set of equations (32), (33) with the initial conditions (34), (35) and the boundary conditions (36), (37) has been solved numerically using a predictor-corrector implicit method[8] for Eq. (32) and determining step by step the evolution of B_Q by Eq. (33). The use of an implicit method does not cause a dramatic increase in the computations, thanks to the quasilinear form of Eq. (32), while it provides the remarkable advantage of numerical stability even below the critical temperature, where other algorithms are much less accurate. In our numerical calculation we have checked that the core condition is indeed very well satisfied along with the evolution of the system over the whole density range. We find that for the fully interacting system at $T \simeq T_c$ the radial distribution function $g(\mathbf{r})$ at $\mathbf{r} = \mathbf{0}$ is at most $\simeq 3 \times 10^{-3}$ and at $T \simeq 1.3\ T_c$ it has already decreased by about an order of magnitude. For $T < T_c$ the situation gets worse, but in every case $g(\mathbf{r} = \mathbf{0})$ does not exceed some 10^{-2} for the temperatures we have investigated. It must be pointed out that below the critical temperature the correct integration of Eqs. (32), (33) is not at all a trivial task, due to the strong nonanalytic behavior inside the coexistence region (see also Section 4) and it is necessary to take great care in the evaluation of the coefficients C, D, K, L because they turn out to be affected by cancellations between extremely divergent quantities.

3. RESULTS ABOVE T_c

We have computed several quantities of the model both above and below T_c. Our results have been compared with those obtained by other theories and by approximants based on extrapolation of perturbative expansions. These approximants can be regarded as the standard reference, since they describe the system with great accuracy. First of all we have located the critical point predicted by the theory. As we have already noted, Eqs. (32), (33) preserve the particle–hole symmetry $\rho \leftrightarrow 1 - \rho$, thus ensuring that the critical density has the correct value $\rho = 1/2$ corresponding to zero magnetic field in the Ising model (we point out that not all theories satisfy this property(9)). The critical temperature determined by the divergence of the isothermal susceptibility χ_T is equal to $kT_c/6J = 0.7553$. This value has to be compared with the "exact" one, $kT_c/6J = 0.7518$, obtained by extrapolation of series expansions (see, e.g., ref. 10); the error is less than 0.5%. In Table I we compare our value of T_c with those

Table I. Critical Temperature for the Three-Dimensional Ising Model on a Simple Cubic Lattice as Given by Several Approximate Theories[a]

Percus–Yevick	ORPA	Bethe	Kikuchi (square)	Kikuchi (cube)	HRT	Exact
0.607	0.660	0.823	0.768	0.763	0.755	0.752

[a] The fourth and fifth values are those from Kikuchi theory using, respectively, a square and a cubic cluster. The value listed for each theory is $kT_c/6J$; mean field theory yields $kT_c/6J = 1$. The Percus–Yevick and ORPA values have been obtained, respectively, from refs. 9 and 19, while the Bethe, Kikuchi, and exact values have been taken from ref. 12.

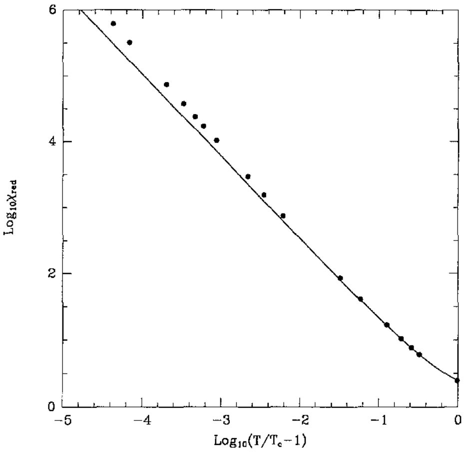

Fig. 1. Log–log plot of the zero-field reduced susceptibility χ_{red} vs. reduced temperature $T/T_c - 1$ close to the critical temperature T_c. Points: HRT. Solid curve: Fisher–Burford approximant.$^{(4)}$

obtained by other liquid-state theories applied to the system in hand, like the ORPA and the Percus–Yevick theories,$^{(6)}$ and by approaches expressly devised for lattice systems, namely the Bethe and the Kikuchi approximations.$^{(11,12)}$ The critical exponent γ has the universal value $\gamma = 1.378$ [see (25)] valid within the class of Ornstein–Zernike approximations of the HRT hierarchy.$^{(1)}$ Figure 1 reports the reduced isothermal susceptibility

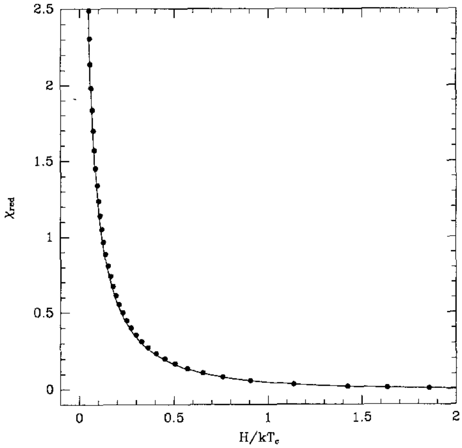

Fig. 2. Reduced susceptibility χ_{red} as a function of the dimensionless magnetic field H/kT_c on the critical isotherm $T = T_c$. Points: HRT. Solid curve: Tarko–Fisher approximant.$^{(5)}$

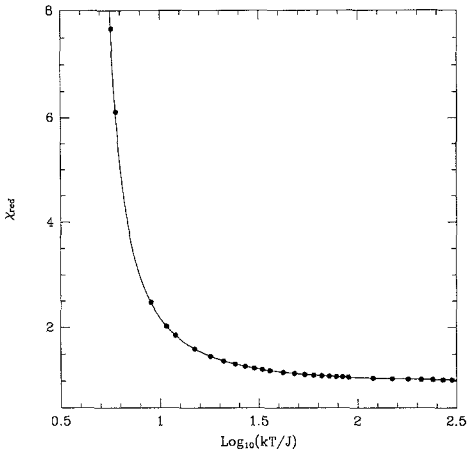

Fig. 3. Zero-field reduced susceptibility χ_{red} vs. dimensionless temperature kT/J for temperatures well above T_c. Points: HRT. Solid curve: Fisher–Burford approximant.$^{(4)}$

$\chi_{\text{red}} = kT\chi_T$ in zero field versus reduced temperature, together with the approximant given by Fisher and Burford.$^{(4)}$ At temperatures very close to T_c a discrepancy can be seen due to the error in our value of γ, the exact value being $\gamma \simeq 1.25$. In Fig. 2 we show our results for the reduced isothermal susceptibility on the critical isotherm compared with the approximant reported by Tarko and Fisher.$^{(5)}$ The value of the critical exponent $\delta = 5$ is again in agreement with (25) and slightly overestimates the exact one ($\delta \simeq 4.8$).

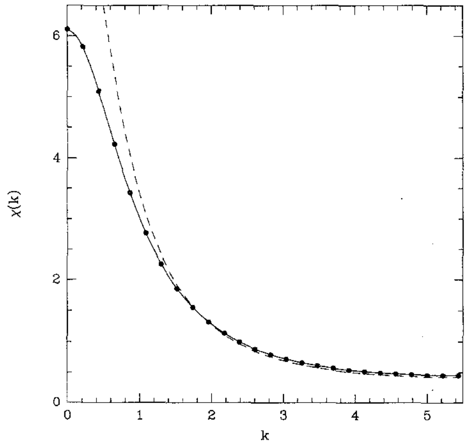

Fig. 4. Structure factor $\chi(\mathbf{k})$ in zero field vs. wavevector modulus k along the direction $k_x = k_y = k_z$ at a reduced temperature $T/T_c = 1.32$. Dashed curve: ORPA. Points: HRT. Solid curve: Fisher–Burford approximant.$^{(4)}$

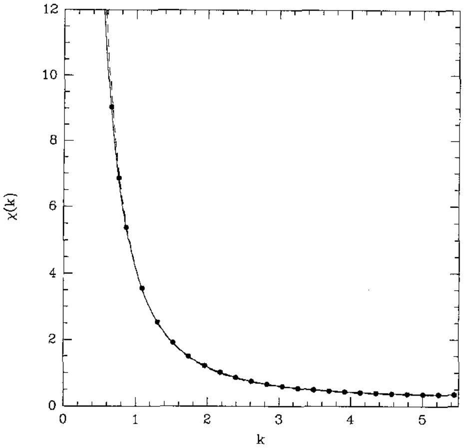

Fig. 5. The same as in Fig. 4, but for $T/T_c = 1.03$. At $\mathbf{k} = \mathbf{0}$, ORPA, HRT, and the Fisher–Burford approximant yield, respectively, $\chi_{\text{red}} = \chi(\mathbf{k} = \mathbf{0}) = 418.4$ (ORPA), $\chi_{\text{red}} = 85.7$ (HRT), $\chi_{\text{red}} = 79.5$ (Fisher–Burford approximant).

Let us consider now the behavior away from the critical point. In Fig. 3 the reduced susceptibility in zero field is plotted over a wide range of temperatures; at high temperature the Curie law typical of a system of noninteracting spins (our reference system) is correctly reproduced. In Figs. 4 and 5 we show the magnetic structure factor $\chi(\mathbf{k})$ in zero magnetic field along the direction $k_x = k_y = k_z$ for two different values of the reduced temperature, together with the results of ORPA(6) and the Fisher–Burford approximant.(4) We have checked that the agreement with the approximant remains unaltered as we consider different directions in the Brillouin zone.

We can then say that above T_c the HRT with the closure relation (22) not only yields nontrivial critical exponents, but it also determines very accurately the nonuniversal behavior of the system and the critical temperature itself. In particular the results for the structure factor show that at temperatures not very close to T_c the Ornstein–Zernike approximation (22) for the present system can be considered very good.(4)

4. RESULTS BELOW T_c

Below the critical temperature the question arises how the theory describes the first-order phase transition taking place in the system. In Figs. 6a and 6b we show the evolution of two magnetization isotherms referring respectively to the cases $T_c < T < T_c^{\text{MF}}$ and $T < T_c$, where T_c^{MF} is the mean field value of the critical temperature given by $kT_c^{\text{MF}}/6J = 1$. The initial value $Q = 0$ corresponds indeed to the mean field approximation, while the final value $Q = -1$ corresponds to the final stage of the integration, in which all the Fourier components of the attractive interaction are taken into account. We see that as long as long-wavelength fluctuations are included in the system the unphysical region where the susceptibility is negative is suppressed: the HRT is thus able to remove the van der Waals loop yielded by mean field theory. For temperatures in the range

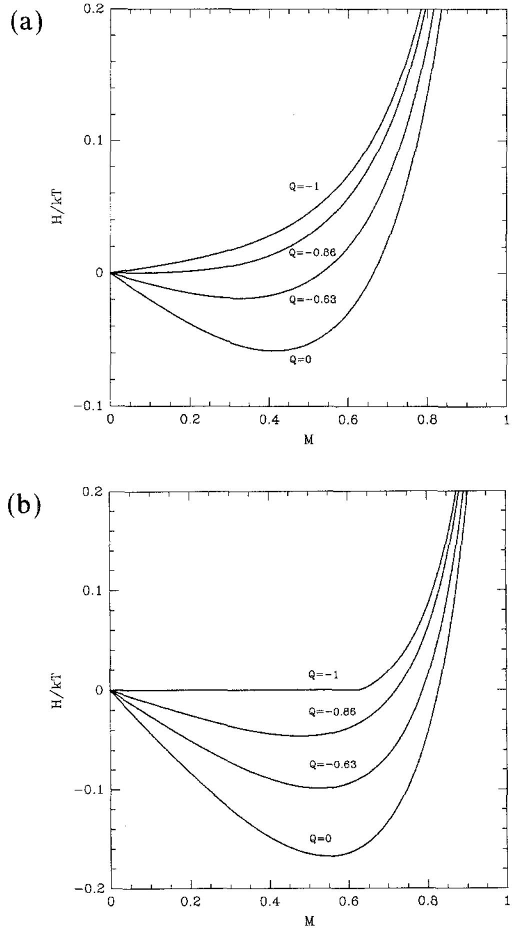

Fig. 6. Evolution of two isotherms above and below T_c as function of the cutoff Q governing the inclusion of the long-range fluctuations. Each curve is labeled by the corresponding value of Q. The curve with $Q = 0$ corresponds to the mean field approximation and that with $Q = -1$ to the fully interacting system. The dimensionless magnetic field H/kT is plotted vs. magnetization M for (a) $T/T_c = 1.1$ and (b) $T/T_c = 0.93$.

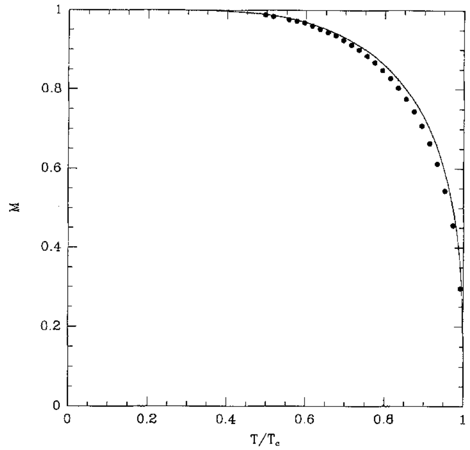

Fig. 7. Spontaneous magnetization M vs. dimensionless temperature T/T_c. Points: HRT. Solid curve: Padé approximant reported by Essam and Fisher.(13)

$T_c < T < T_c^{\mathrm{MF}}$ the loop disappears altogether, but below T_c it is replaced by a flat region where $H=0$ and $M \neq 0$, which is nothing but the coexistence region. For any given temperature $T < T_c$ the amplitude of the coexistence region can be determined unambiguously as the extension of the flat portion of the isotherm and this gives the value of the spontaneous magnetization M. In Fig. 7 the spontaneous magnetization as a function of temperature (i.e., the coexistence curve) is compared with a Padé approximant for the same quantity.(13) The critical exponent β is again found to be in agreement with the value $\beta = 0.345$ [see (25)] obtained from the asymptotic analysis and the scaling laws. The fact that the HRT has the correct scaling regime both above and below the critical temperature is clearly shown in Fig. 8: here we plot the scaling function $h(x)$ which determines the equation of state in the critical region according to the expression $H/kT_c = M^\delta h(t/M^{1/\beta})$, where t is the reduced temperature $(T-T_c)/T_c$. One can see that for small enough t the points from different isotherms all fall on the same curve; we remark that the two branches for $t>0$ and $t<0$ match very well each other. In the figure the approximants for $h(x)$ given by Gaunt and Domb(14),[3] is also shown; the discrepancy

---

[3] Actually the results reported by Gaunt and Domb refer to the body-centered cubic lattice; the lattice structure affects the nonuniversal features of the scaling function, namely the value x_0 such that $h(x_0)=0$ and the value $h_0 = h(x=0)$. Those values in turn depend, respectively, on the amplitudes of the spontaneous magnetization and the critical isotherm in the critical region. In order to take into account the influence of the lattice, the approximant has been properly rescaled; the amplitudes for the simple cubic lattice have been taken from ref. 10 (spontaneous magnetization) and from ref. 20 (critical isotherm).

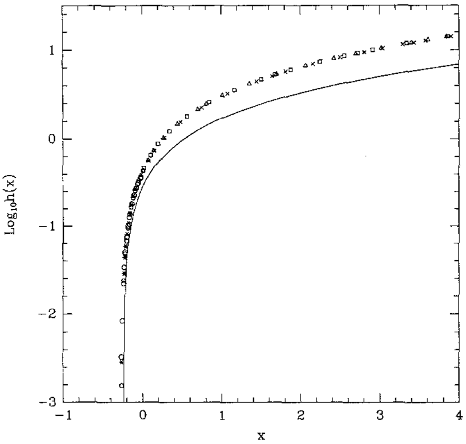

Fig. 8. Semilog plot of the scaling function $h(x)$. The abscissa x is equal to $t/M^{1/\beta}$, while $h(x)$ is given by $H/(kT_cM^{\delta})$; t is the reduced temperature $T/T_c - 1$, and β and δ are the usual critical exponents. The various points refer to HRT results for different isotherms. Squares: $t = 4.38 \times 10^{-5}$. Triangles: $t = 7.02 \times 10^{-5}$. Crosses: $t = 2.03 \times 10^{-4}$. Asterisks: $t = -6.21 \times 10^{-5}$. Pentagons: $t = -1.95 \times 10^{-4}$. Circles: -3.27×10^{-4}. Solid line: Gaunt and Domb approximant.(14)

between the two curves increases on increasing x, due to the error in the critical exponent γ. [We recall that one has $h(x) \sim x^{\gamma}$ for large x.]

The behavior across the coexistence curve is investigated in more detail in Fig. 9, where we report the inverse reduced susceptibility as a function of the magnetization compared with the low-temperature expansion data(15) in a small interval around the boundary of the coexistence region at a temperature $kT/6J = 0.47$. We see that across this region the inverse susceptibility varies very steeply but remains nevertheless continuous and thus goes to zero even when we approach the coexistence curve from outside, as happens in the case of XY and Heisenberg models. This gives rise to an infinite value of the zero-field susceptibility for every $T \leqslant T_c$, in contrast with the true behavior of the system. We recall that the only approximation made in our calculation is the closure relation (22) for $\mathscr{C}_Q(\mathbf{k})$, which must then be responsible for this spurious result. Moreover, since the onset of the first-order phase transition is governed by the long-wavelength fluctuations, we expect that the relevant information entering in approximation (22) is the behavior of $\mathscr{C}_Q(\mathbf{k})$ in the small-k limit [see Eq. (24)]. We notice, however, that the dimensionality d of the system plays an important role as well, and for $d > 4$ a more realistic description of the first-order transition is found using the same kind of closure.(16)

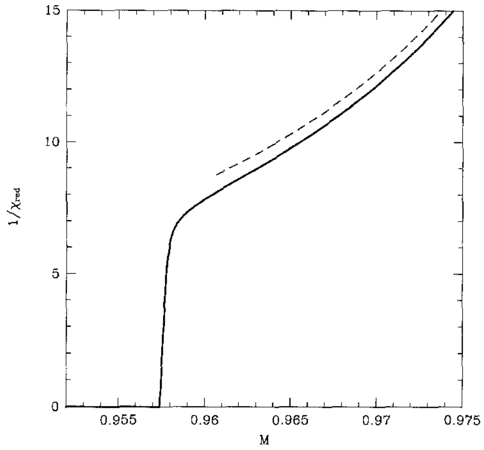

Fig. 9. Inverse reduced susceptibility $1/\chi_{\rm red}$ vs. magnetizeation M across the boundary of the coexistence region at a temperature $kT/6J = 0.47$. Solid curve: HRT. Dashed curve: data from the low-temperature expansion.(15)

Although our choice for $\mathscr{C}_Q(\mathbf{k})$ is not flexible enough to model the correlations in the coexistence region, it is nevertheless interesting to observe that the nonanalytic behavior of the theory in this region strongly affects the coefficient $B_Q(\rho, T)$ in Eq. (22), which fixes the curvature of the direct correlation function. In fact putting in the core condition (26) $E_Q(\rho, T) = 0$ and solving with respect to $B_Q(\rho, T)$ would yield for the completely interacting system the temperature-independent value

$$B_Q(\rho, T) = -6W_0/\rho(1-\rho) \simeq -1.5/\rho(1-\rho)$$

where $W_0 \simeq 0.252731$ is the Watson function in three dimensions,(7) $W(x)$ for $x = 0$. However, in the coexistence region the limiting procedure with respect to Q does not commute with the integration in Eq. (26) and this gives rise to a completely different behavior for B_Q: actually, it can be seen that inside the coexistence region B_Q does not depend on density; moreover, it increases very rapidly in absolute value on decreasing temperature, causing the direct correlation function to be very steep near $\mathbf{k} = \mathbf{0}$. This seems to indicate that as Q evolves, our direct correlation function tries as much as possible to reproduce the discontinuity at $\mathbf{k} = \mathbf{0}$ inside the coexistence region shown by the true direct correlation function of the fully interacting system, which vanishes precisely at $\mathbf{k} = \mathbf{0}$ but is finite in the $\mathbf{k} \to \mathbf{0}$ limit.(17) Of course, the attempt must ultimately fail, since the closure relation (22) for $\mathscr{C}_Q(\mathbf{k})$ is always a continuous function of $\mathbf{k}$.

5. CONCLUSIONS

We have applied the HRT with an Ornstein–Zernike closure to the three-dimensional Ising model both above and below T_c. Our results show that the theory describes with considerable accuracy several nonuniversal properties of the system; in particular the value of the critical temperature differs from the exact one by less than 0.5% and so it is one of the most accurate between those yielded by approximate theories. At the same time the behavior in the critical region is much more realistic than in standard approximations, the critical exponents are not trivial, and the scaling regime is correct. The critical exponents are not very accurate, however, and this can be traced to the adopted closure, which implies that $\eta = 0$. In order to go beyond this approximation one should take into account the second equation of the hierarchy. A particularly interesting feature of the HRT is that it retains the convexity of the free energy even below T_c, suppressing the van der Waals loop and replacing it with the plateau typical of the coexistence region as long as long-wavelength fluctuations are included in the system. The main shortcoming of our approach is that the simple from (22) assumed for the direct correlation function cannot describe the correlations inside the coexistence region in a realistic way and for a three-dimensional system it fails to reproduce the discontinuous jump of the inverse susceptibility across the coexistence curve. In fact, our approximation gives an infinite value of the susceptibility on the coexistence curve. This behavior would be appropriate for a system whose order parameter has more than one component. It might thus be interesting to apply the HRT with the same kind of Ornstein–Zernike approximation used here to other magnetic models, like *XY* or Heisenberg models or even Ising systems with competing interactions.(18)

ACKNOWLEDGMENTS

This work has been supported by Ministero dell'Università e della Ricerca Scientifica, by Istituto Nazionale di Fisica della Materia and by Consiglio Nazionale delle Ricerche under Progetto Finalizzato "Sistemi Informatici e Calcolo Parallelo".

APPENDIX

In this Appendix Eqs. (32), (33) are derived in detail starting from Eq. (27) for the evolution of the free energy $\mathcal{A}_Q$ and from the core condition (29).

Let us consider again Eq. (27):

$$\frac{\partial \mathscr{A}_Q}{\partial Q} = -\frac{1}{2}F(Q)\left[\log\left(1-\frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})}\right)\bigg|_{\gamma(\mathbf{k})=Q} + \log\left(1-\frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})}\right)\bigg|_{\gamma(\mathbf{k})=-Q}\right] \tag{A1}$$

If we use the explicit expression of $\tilde{\Phi}(\mathbf{k})$,

$$\tilde{\Phi}(\mathbf{k}) = \lambda\gamma(\mathbf{k}), \qquad \lambda = \frac{6w}{kT} \tag{A2}$$

and the closure relation

$$\mathscr{C}_Q(\mathbf{k}) = E_Q + B_Q[1-\gamma(\mathbf{k})] \tag{A3}$$

then Eq. (A1) becomes

$$\frac{\partial \mathscr{A}_Q}{\partial Q} = -\frac{1}{2}F(Q)\log\left[1-\frac{\lambda Q^2(2B_Q+\lambda)}{(E_Q+B_Q)^2-B_Q^2Q^2}\right] \tag{A4}$$

We now differentiate Eq. (A4) twice with respect to ρ and take account of the thermodynamic consistency requirement

$$\mathscr{C}_Q(\mathbf{k}=0) = E_Q(\rho, T) = \frac{\partial^2 \mathscr{A}_Q}{\partial \rho^2} \tag{A5}$$

to get

$$\frac{\partial E_Q}{\partial Q} = -\frac{1}{2}Q^2F(Q)\frac{\partial^2 v_Q}{\partial \rho^2} \tag{A6}$$

where we have defined

$$v_Q = \frac{1}{Q^2}\log\left[1-\frac{\lambda Q^2(2B_Q+\lambda)}{(E_Q+B_Q)^2-B_Q^2Q^2}\right] \tag{A7}$$

The above relation can be inverted to express E_Q in terms of v_Q and B_Q:

$$E_Q = Q\chi - B_Q \tag{A8}$$

where χ is defined as

$$\chi = \left[\frac{\lambda(2B_Q+\lambda)}{1-e^{Q^2v_Q}} + B_Q^2\right]^{1/2} \tag{A9}$$

Using (A8) in the left-hand side of (A6) yields

$$Q^2 \frac{\partial^2 v_Q}{\partial \rho^2} = C'(Q, v_Q, B_Q) \frac{\partial v_Q}{\partial Q} + D'(Q, v_Q, B_Q) \tag{A10}$$

where we have set

$$C'(Q, v_Q, B_Q) = -\frac{\lambda Q^3 (2B_Q + \lambda) e^{Q^2 v_Q}}{F(Q)(1 - e^{Q^2 v_Q})^2 \chi} \tag{A11}$$

$$D'(Q, v_Q, B_Q) = -\frac{2}{F(Q)} \left[D_1(Q, v_Q, B_Q) + D_2(Q, v_Q, B_Q) \frac{\partial B_Q}{\partial Q} \right] \tag{A12}$$

and

$$D_1(Q, v_Q, B_Q) = \frac{\lambda Q^2 (2B_Q + \lambda) e^{Q^2 v_Q}}{(1 - e^{Q^2 v_Q})^2 \chi} v_Q + \chi \tag{A13}$$

$$D_2(Q, v_Q, B_Q) = Q \frac{1}{\chi} \left(\frac{\lambda}{1 - e^{Q^2 v_Q}} + B_Q \right) - 1 \tag{A14}$$

We now turn to the core condition (29),

$$\begin{aligned} &\int_{\Omega_Q} \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{1}{[\mathscr{C}_Q(\mathbf{k}) - \tilde{\Phi}(\mathbf{k})]^2} \frac{d\mathscr{C}_Q}{dQ}(\mathbf{k}) + \int_{\mathscr{B} - \Omega_Q} \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{1}{\mathscr{C}_Q^2(\mathbf{k})} \frac{d\mathscr{C}_Q}{dQ}(\mathbf{k}) \\ &\quad = F(Q) \left[\left. \frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})[\mathscr{C}_Q(\mathbf{k}) - \tilde{\Phi}(\mathbf{k})]} \right|_{\gamma(\mathbf{k}) = Q} + \left. \frac{\tilde{\Phi}(\mathbf{k})}{\mathscr{C}_Q(\mathbf{k})[\mathscr{C}_Q(\mathbf{k}) - \tilde{\Phi}(\mathbf{k})]} \right|_{\gamma(\mathbf{k}) = -Q} \right] \end{aligned} \tag{A15}$$

Again we use in (A15) the explicit expressions (A2), (A3) for $\tilde{\Phi}(\mathbf{k})$ and $\mathscr{C}_Q(\mathbf{k})$ and recall that the region Ω_Q has been defined in Section 2 by $|\gamma(\mathbf{k})| > -Q$, $-1 \leqslant Q \leqslant 0$, to get

$$R(Q, v_Q, B_Q) \frac{\partial E_Q}{\partial Q} + S(Q, v_Q, B_Q) \frac{\partial B_Q}{\partial Q} = H(Q, v_Q, B_Q) \tag{A16}$$

where H, R, S are given by

$$H(Q, v_Q, B_Q) = \frac{2F(Q)(1 - e^{Q^2 v_Q})^2 \chi}{\lambda Q (2B_Q + \lambda) e^{Q^2 v_Q}} \tag{A17}$$

$$R(Q, v_Q, B_Q) = 2Q^2 \chi^2 I + 2(B_Q + \lambda)^2 J + M \tag{A18}$$

$$S(Q, v_Q, B_Q) = 2Q^2 \chi^2 I + 2(B_Q + \lambda)(B_Q + \lambda - 2Q\chi) J + N \tag{A19}$$

and I, J, M, N are the following integrals:

$$I = \int_{-1}^{Q} dx\, F(x) \frac{1}{[Q^2\chi^2 - (B_Q + \lambda)^2 x^2]^2} \tag{A20}$$

$$J = \int_{-1}^{Q} dx\, F(x) \frac{x^2}{[Q^2\chi^2 - (B_Q + \lambda)^2 x^2]^2} \tag{A21}$$

$$M = \int_{Q}^{-Q} dx\, F(x) \frac{1}{(Q\chi - B_Q x)^2} \tag{A22}$$

$$N = \int_{Q}^{-Q} dx\, F(x) \frac{1 - x}{(Q\chi - B_Q x)^2} \tag{A23}$$

If we combine Eqs. (A6) and (A16), we obtain

$$\frac{\partial B_Q}{\partial Q} = \frac{1}{S(Q, v_Q, B_Q)} \left[H(Q, v_Q, B_Q) + \frac{1}{2} R(Q, v_Q, B_Q)\, Q^2 F(Q) \frac{\partial^2 v_Q}{\partial \rho^2} \right] \tag{A24}$$

Equation (A24) can be used in (A12) to eliminate $\partial B_Q/\partial Q$ in favor of $\partial^2 v_Q/\partial \rho^2$. Then Eq. (A10) becomes

$$Q^2 \frac{\partial^2 v_Q}{\partial \rho^2} = C(Q, v_Q, B_Q) \frac{\partial v_Q}{\partial Q} + D(Q, v_Q, B_Q) \tag{A25}$$

with the definitions

$$C(Q, v_Q, B_Q) = \frac{C'S}{S + D_2 R} \tag{A26}$$

$$D(Q, v_Q, B_Q) = -\frac{2}{F(Q)} \frac{D_1 S + D_2 H}{S + D_2 R} \tag{A27}$$

where we have understood the dependence on Q, v_Q, B_Q in the above-defined quantities C', H, D_1, D_2, R, S. Finally, we use (A25) in (A24) to obtain

$$\frac{\partial B_Q}{\partial Q} = K(Q, v_Q, B_Q) \frac{\partial v_Q}{\partial Q} + L(Q, v_Q, B_Q) \tag{A28}$$

where

$$K(Q, v_Q, B_Q) = \frac{1}{2} F(Q) \frac{C'R}{S + D_2 R} \tag{A29}$$

$$L(Q, v_Q, B_Q) = \frac{H - D_1 R}{S + D_2 R} \tag{A30}$$

Equations (A25) and (A28) are the quasilinear equations for v_Q and B_Q reported in Section 2. The definitions (A26), (A27), (A29), and (A30) give the explicit expressions of the coefficients C, D, K, and L.

REFERENCES

1. A. Parola and L. Reatto, *Phys. Rev. A* **31**:3309 (1985).
2. A. Meroni, A. Parola, and L. Reatto, *Phys. Rev. A* **42**:6104 (1990).
3. K. Huang, *Statistical Mechanics* (Wiley, New York, 1987).
4. M. E. Fisher and R. J. Burford, *Phys. Rev.* **156**:583 (1967).
5. H. B. Tarko and M. E. Fisher, *Phys. Rev. B* **11**:1217 (1975).
6. J. P. Hansen and I. R. McDonald, *Theory of Simple Liquids* (Academic Press, London, 1986).
7. S. Katsura, T. Morita, S. Inawashiro, T. Horiguchi, and Y. Abe, *J. Math. Phys.* **12**:892, 895, 981 (1971).
8. W. F. Ames, *Numerical Methods for Partial Differential Equations* (Academic Press, New York, 1977).
9. D. Levesque and L. Verlet, *Phys. Lett.* **11**:36 (1964).
10. C. Domb, in *Phase Transition and Critical Phenomena*, Vol. 3, C. Domb and M. S. Green, eds. (Academic Press, London, 1974).
11. R. Kikuchi, *Phys. Rev.* **81**:988 (1951).
12. D. M. Burley, in *Phase Transition and Critical Phenomena*, Vol. 2, C. Domb and M. S. Green, eds. (Academic Press, London, 1972).
13. J. W. Essam and M. E. Fisher, *J. Chem. Phys.* **38**:802 (1963).
14. D. S. Gaunt and C. Domb, *J. Phys. C* **3**:1442 (1970).
15. G. Bhanot, M. Creutz, and J. Lacki, *Phys. Rev. Lett.* **69**:1841 (1992).
16. A. Parola, D. Pini, and L. Reatto, *Phys. Rev. E*, to be published.
17. M. E. Fisher, *Rep. Prog. Phys.* **30**:615 (1967).
18. D. Mukamel and S. Krinsky, *Phys. Rev. B* **13**:5078 (1976).
19. B. Carazza, A. Parola, L. Reatto, and M. Tau, *Physica A* **116**:207 (1982).
20. D. S. Guant, *Proc. Phys. Soc.* **92**:150 (1967).
21. G. Stell, J. L. Lebowitz, S. Baer, and W. Theumann, *J. Math. Phys.* **7**:1532 (1966).